COHERENTLY STIMULATED BRILLOUIN SPECTROSCOPY

By Joel N. Johnson

A Dissertation

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Dedication

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Preface

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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 changes the mechanical energy of the material. 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 with light by retrieving 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. Here I offer a brief overview of notable optomechanical phenomena then devote the remainder of this chapter to a more detailed description of the specific interactions that play a role in my research. 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 . 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 . This technique becomes especially effective when combined with nanoparticle-enhanced absorption, allowing for dramatically increased absorption in ultra-localized zones within the body.

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 (λ) 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. [1]

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.

Brillouin scattering, around which much of my work 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.

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.

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

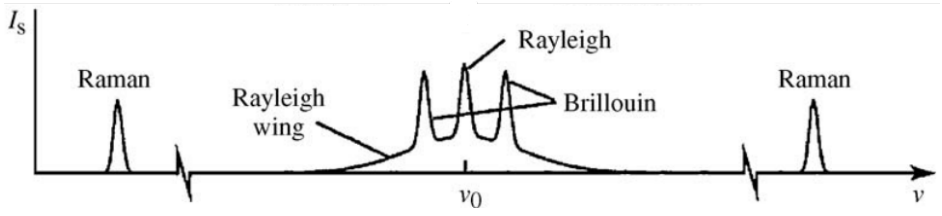

Figure 1.1: Relative domains of typical frequency shifts for Rayleigh, Rayleigh-wing, Brillouin, and Raman scattering.

understanding their common frequency domains. Moreover, it serves as a reminder of the rich continuum of material behavior and responses that affect light scattering as opposed to the distinct categories we ascribe for convenience. This is a core concept of my work.

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.

The final category of optomechanical interactions I will note 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.[2] 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[3], just one year after the first demonstration of the laser by American physicist Theodor Maiman.[4] These nonlinear effects provide the foundation for a range of technologies, including high-speed optical communication systems, frequency converters, and lasers for materials processing.

Also included within 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 an optically induced strain. These effects are utilized in designing optical modulators, tunable photonic devices, and

smart materials that respond to light .

In the remainder of this chapter I further describe the specific optomechanical phenomena that pertain to the research presented in this document: Brillouin scattering, electrostriction as it pertains to the SBS process, and Raman scattering.

1.1 Light Scattering

Light scattering involves the redirection of light as a result of interactions with the constituent particles or molecules within a material medium. In every case, light scattering occurs because of variations in the material's optical properties. To understand why, envision a material with completely uniform particles—spatially and temporally consistent, or in other words, perfectly homogeneous. Figure 1.2 shows an incident optical plane wave encountering a segment of such a material, denoted δz, containing a volume element δV_1. For any given incident wavelength λ and any non-zero scattering angle θ at volume δV_1, there exists a corresponding volume element δV_2, located a distance $\frac{\lambda}{2\sin\theta}$ apart, which scatters light at the same angle θ. The scattered waves from δV_1 and δV_2 would be out of phase by $\frac{\lambda}{2}$, leading to perfect destructive interference and no resultant scattered field. Thus, to achieve observable scattering, the material must possess inhomogeneities, allowing for variations in the optical properties between neighboring volumes. Fortunately, perfect homogeneity is not characteristic of real materials; all matter undergoes thermodynamic fluctuations at any temperature above absolute zero, and quantum fluctuations are inherent even at the ground state.

I now begin with a theoretical description of spontaneous light scattering as a result of thermodynamic fluctuations, presented in Boyd Nonlinear Optics.[2] This foundation will serve as a framework for understanding light scattering as specifically resulting from pressure variations (Brillouin scattering) as opposed to density variations (Rayleigh scattering). Later I will treat the case of higher-intensity SBS. Ultimately I will build upon this theoretical basis to derive the coupled-wave equations of the Coherent Anti-Stokes Brillouin Spectrometer (CABS), a novel instrument which underpins many of my results. Let us build a theoretical description of light scattering considering thermodynamic fluctuations as the origin of the scattering process.

1.2 Spontaneous Brillouin Scattering

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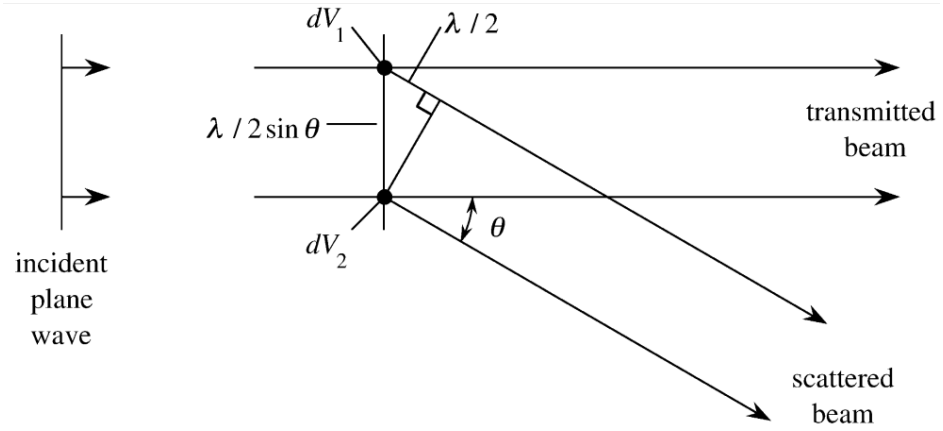

Figure 1.2

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1.3 Stimulated Brillouin Scattering

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1.4 Phase-matching

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1.5 Brillouin Gain of Materials

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1.6 Raman Scattering

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1.7 Raman-like Brillouin Modes

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Chapter 2

Foundational Experimental Techniques and Instrumentation

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2.1 Photonic Experimental Techniques

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2.1.1 Control of Light in Photonic Systems

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2.1.2 Photonic Devices and Diagrams

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2.1.3 Selection and Isolation of Signals

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2.1.4 Heterodyne Detection and the Local Oscillator

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2.1.5 Optical Loss in a Photonic Systems

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2.1.6 Free Space Optics and Beam Alignment

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2.1.7 Specialized Optical Fibers

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2.2 Optical Instrumentation

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2.3 Electronic Instrumentation

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2.4 Noise and Background Handling

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2.5 Custom Software

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2.5.1 Description of Python Script for CABS Data Collection

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2.5.2 Description of Plotting Data in Go Program

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Chapter 3

Manuscript I: Laser cooling of traveling wave phonons in an optical fiber

3.1 Abstract

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3.2 Optomechanical Cooling and Heating

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3.3 Cooling Platform: CS_2-Liquid Core Optical Fiber

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3.3.1 Optomechanical Properties

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3.3.2 Fabrication

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3.3.3 Fabrication Iterative Refinement

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3.4 Intention of the Pump-Probe Experiment

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3.5 Experimental Setup

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3.5.1 Main Experiment

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3.5.2 Pump-Probe Experiment

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3.6 Results

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3.6.1 Main Experiment Results

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3.6.2 Pump-Probe Experiment Results

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3.7 Discussion

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3.7.1 Application to Ground State Cooling

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3.7.2 Standardized Cooling Metric

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3.7.3 Tapered chalcogenide Photonic Crystal Fiber: Max Plank Results

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Chapter 4

Manuscript II: A coherently stimulated phonon spectrometer

Joel N. Johnson[1,2], Nils T. Otterstrom[3], Peter T. Rakich[4], Ryan O. Behunin[1,2]

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4.1 Abstract

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4.2 Introduction

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[3] Sandia National Laboratory, 1515 Eubank Blvd SE, Albuquerque, NM 87123, USA
[4] Department of Applied Physics, Yale University, New Haven, CT 06520, USA

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4.3 Instrument Design

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4.3.1 Design of instrument

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4.3.2 Sensitivity Measurements

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4.4 Theory

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4.4.1 Coupled Wave Equations

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4.4.2 Phase-matching bandwidth

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4.5 Results

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4.5.1 Fiber-Coupled: UHNA3

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4.5.2 Free-Space: CS_2

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comparison to stimulated brillouin and spontaneous brillouin?

4.5.3 Phase-Matching in Small L Regime

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4.6 Discussion

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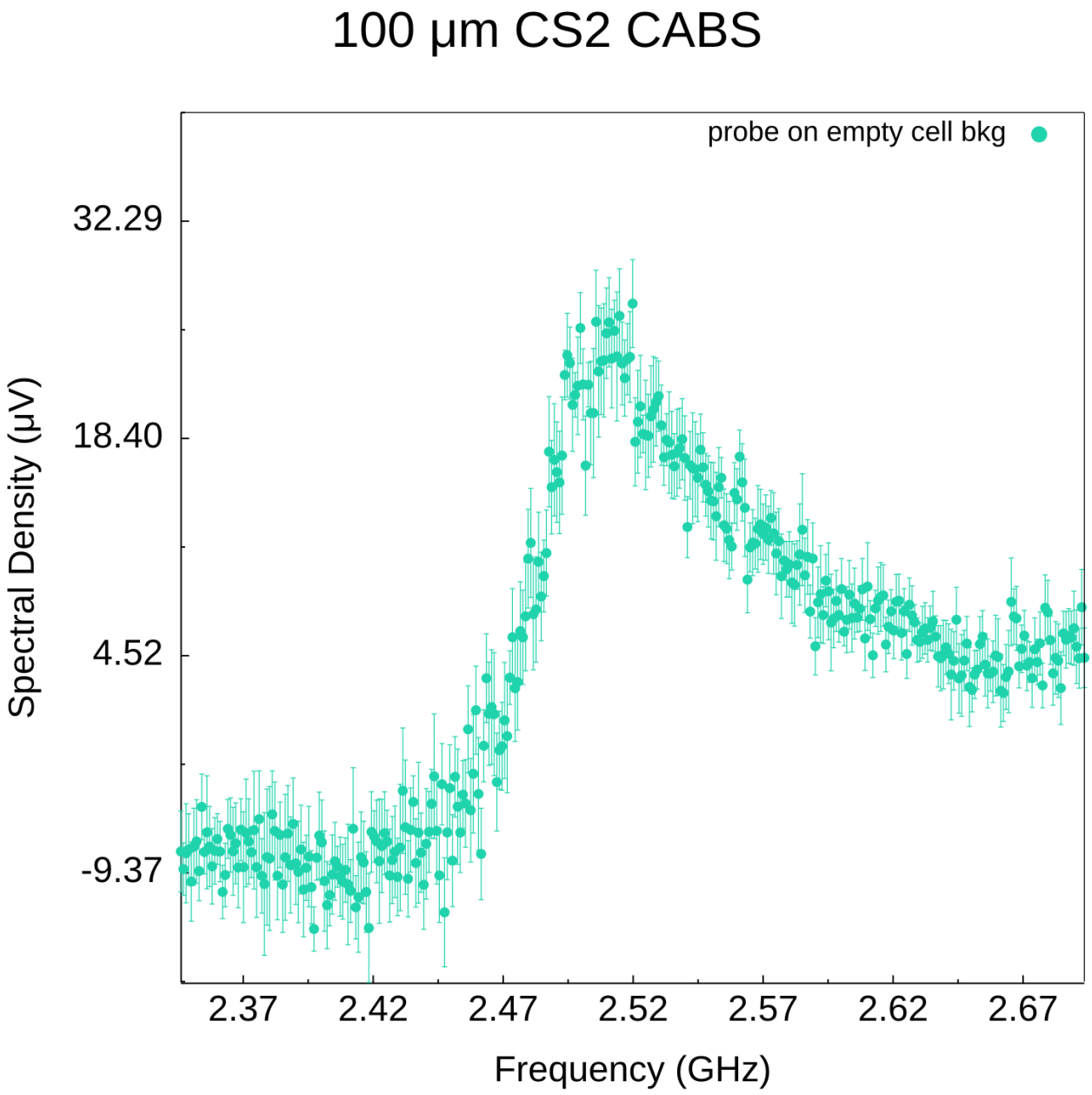

Figure 4.1: CABS measurement of 100um of CS2.

Chapter 5

Manuscript III: Brillouin-induced Raman modes

5.1 Abstract

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5.2 Introduction

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5.3 Methods

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5.4 Results

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5.5 Discussion

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rutrum.

Table 5.1: Table caption.

	Parameter	Value	Description
Lookup Variables	lat	-85°–85°	Latitude (35 bins in 5° increments)
	ALBEDO	0.05–0.225	Bolometric albedo (6 bins in 0.035 increments)
	SLOPE	0°–90°	Surface slope (19 bins in 5° increments)
	SLOAZI	0°–360°	Surface azimuth (19 bins in 20° increments)
	DELLS	4°	L_s step size (90 bins spanning 0°–360°)
Thermal Parameters	EMISS	0.96	Emissivity
	thick	0.05	Upper layer thickness [m]
	DENSITY	1100	Upper layer density [kg/m^3]
	DENS2	1800	Lower layer density [kg/m^3]
	lbound	18	Interior heat flow [mW/m^2]
	PhotoFunc	0.045/albedo	Photometric function (Keihm-style)
Temperature-dependent parameters	SphUp0/SphLo0	602.88098583	Specific heat capacity expressed as 4th-order polynomial ($c0 + c1 \cdot T + c2 \cdot T^2 + c3 \cdot T^3$)
	SphUp1/SphLo1	235.98988249	
	SphUp2/SphLo2	-29.59742178	
	SphUp3/SphLo3	-3.78707193	
	ConUp0	0.00133644	Upper layer conductivity expressed as 4th-order polynomial ($c0 + c1 \cdot T + c2 \cdot T^2 + c3 \cdot T^3$)
	ConUp1	0.00073150	
	ConUp2	0.00033250	
	ConUp3	0.00005038	
	ConLo0	0.00634807	Lower layer conductivity expressed as 4th-order polynomial ($c0 + c1 \cdot T + c2 \cdot T^2 + c3 \cdot T^3$)
	ConLo1	0.00347464	
	ConLo2	0.00157938	
	ConLo3	0.00023930	
Model Setup Parameters	body	Moon	Target body
	k_style	Moon	Conductivity style (Moon for airless bodies)
	LKofT	T	Temperature-dependent conductivity
	FLAY	0.01	First layer thickness [m]
	RLAY	1.3	Layer thickness multiplier
	N1	26	Number of layers
	N24	288	Timesteps per day (5 min steps)
	DJUL	0	Start date

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Chapter 6

Discussion & Future Work

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Appendix A

Acronyms

SBS Stimulated Brillouin Scattering

CABS the Coherent Anti-Stokes Brillouin Spectrometer

Appendix B

Code

B.1 Python Code for CABS Data Collection

B.2 Plotting Data In Go Program

Appendix C

Supplementary Information for Chapter 3: Manuscript I

C.1 Data

Appendix D

Supplementary Information for Chapter 4: Manuscript II

D.1 Equal Contribution of Pump, Stokes, and Probe Optical Fields

Equation **??** 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.

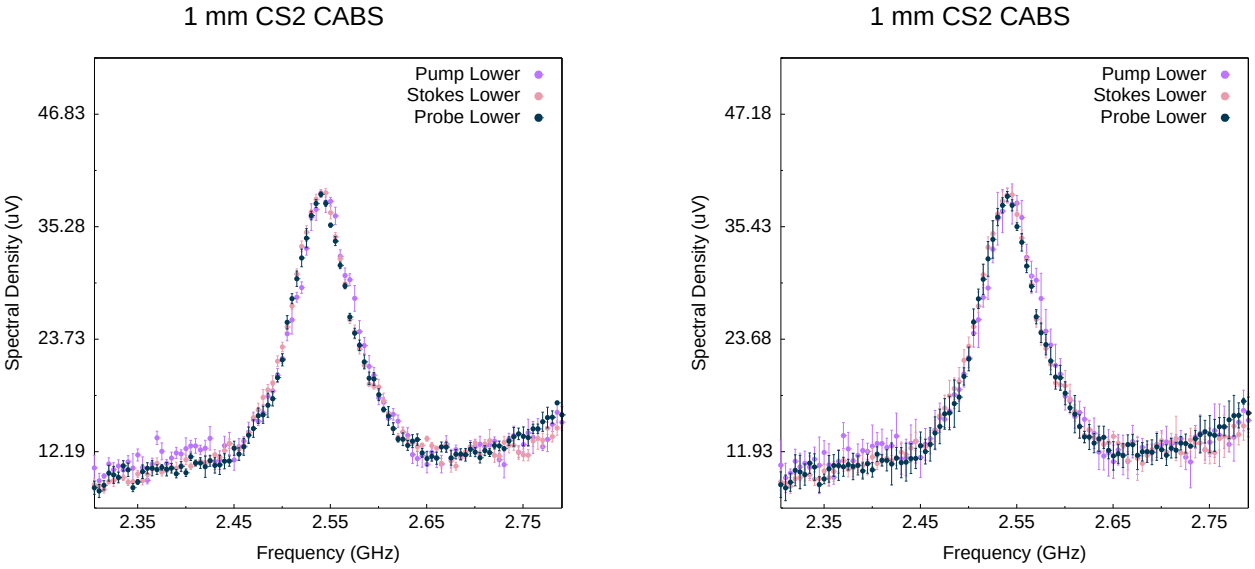
To test this, we conducted a controlled experiment with a 1 mm carbon disulfide (CS_2) sample. For each measurement, one of the three source powers (Pump, Stokes, or Probe) was systematically reduced by 75% while holding the others constant and ensuring consistent experimental conditions across trials. Table D.1 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^3)**
Pump Lower	19.190	32.210	54.560	3.372×10^4
Stokes Lower	76.600	8.020	54.650	3.359×10^4
Probe Lower	76.600	32.530	13.480	3.359×10^4

Table D.1: Power values for each source (Pump, Stokes, Probe) across the three measurements, with the multiplicative total power for each setup.

Figure D.1a displays the average results from these three measurements, plotted with error bars representing one standard deviation of the mean. For increased certainty, Figure D.1b 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 **??** for predicting Signal power across a range of power distributions within practical settings.



(a) Signal power contributions with error bars representing one standard deviation of the mean for each measurement.

(b) Signal power contributions with error bars extended to two standard deviations of the mean for each measurement.

Figure D.1: Comparison of Signal power contributions with error bars representing one (a) and two (b) standard deviations of the mean for each measurement.

D.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 instrumdent 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 scatterd power at greater lengths as well (greater than 1 meter). This is because the reduction in scattered power from the breakdown of phase-matching relaxation at greater lengths is perfectly counter-balanced by the quadradic dependence on length in the overall scatterd power, as seen in Eq. **??**. 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 paramters. 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.[2] In this low-gain regime, any scattered Stokes light is spontanously 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 $W^{-1}m^{-1}$ ($G_B = \frac{g}{A_{eff}}$), 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[5] as

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

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. 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,

$$P_S = \frac{1}{4} G_B P_P L \hbar \omega_S \Gamma_B (\bar{n} + 1). \tag{D.2}$$

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

$$e^{\frac{\hbar\Omega_B}{k_b T}} \approx 1 + \frac{\hbar\Omega_B}{k_b T} \tag{D.3}$$

to be a good approximation. We thus find that

$$(\bar{n} + 1) \approx \bar{n} \approx \frac{k_b T}{\hbar\Omega_B}. \tag{D.4}$$

Inserting this reduced quantity into Eq. D.2, we arrive at a convenient expression for the scattered power of the Stokes spontaneous Brillouin scattering process,

$$P_{S,\,SponBS} = \frac{G_B P_P L \omega_S \Gamma_B k_b T}{4\Omega_B}. \tag{D.5}$$

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 [6] for the complementary forward scattering processe. While the two scenarios—our backward scattering geometry versus the forward scattering geometry discussed by Kharel et al.—differ in directionality, the underlying physics of light coupling to thermally excited acoustic modes is the same and reflects the fundamental similarity in how thermal phonons mediate the interaction between optical fields in the low-gain (spontaneous) regime.

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 $20 < G < 25$, [5] whereas for typical lengths of single mode fiber it can be lower [7] 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 [5]

$$R = \frac{\langle |E_S|^2 \rangle}{\langle |E_P|^2 \rangle} = \frac{Y}{\sqrt{\pi}} \frac{e^G}{G^{\frac{3}{2}}}, \tag{D.6}$$

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,

$$P_{S,\,StimBS} = \frac{G_B P_P L \omega_S \Gamma_B k_b T}{4\sqrt{\pi}\Omega_B} \frac{e^G}{G^{\frac{3}{2}}} \tag{D.7}$$

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 Eq. D.7 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. [2] To account for pump depletion, we numerically solve the transendental equation derived in Boyd's Nonlinear Optics which describes the effects of pump depletion, given here in terms of power as

$$P_S(L) = \frac{P_S(0) x (1-x)}{e^{G_B P_P(0) L (1-x)} - x}, \tag{D.8}$$

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

$$P_S(0) = xP_P(0). \tag{D.9}$$

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 Fig. D.2, along with the analytical solutions derived previously for the low-gain (spontaneous) regime and our coherently stimulated Brillouin spectrometer given by Eq. **??**. System parameters used to generate the plot for each of the three processes are provided in Tables D.2 and D.3. 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.

Coherently Stimulated Brillouin Scattering Process Model System Parameters

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

Table D.2: Parameters relevant to the coherently stimulated backward Brillouin scattering process for the example 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.

Spontaneous and Stimulated Scattering Process Model System Parameters

G_B	P_P	$P_{S,seed}$	n	λ_P	Γ_B	k_B	T	Ω_B
0.6 $W^{-1}m^{-1}$	1 W	1 pW	1.48	1549 nm	$2\pi \cdot 80$ MHz	1.38×10^{-23} J/K	295 K	$2\pi \cdot 9.18$ GHz

Table D.3: 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, ω is the optical angular frequency, Γ_B is the acoustic damping rate, k_B is Boltzmann's constant, T is the temperature, and Ω_B is the acoustic angular frequency.

At lengths beyond a centimeter, the phase-matching relaxation of the coherently stimulated process begins to break down, and the specific choice in pump and probe detuning becomes critical. This corresponds to a narrowing of the $sinc^2$ function given in Eq. **??**. The scattered power beyond this length rises and falls according to the oscillations of the $sinc^2$ function far from the origin. As length increases continuously beyond 1 meter, 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

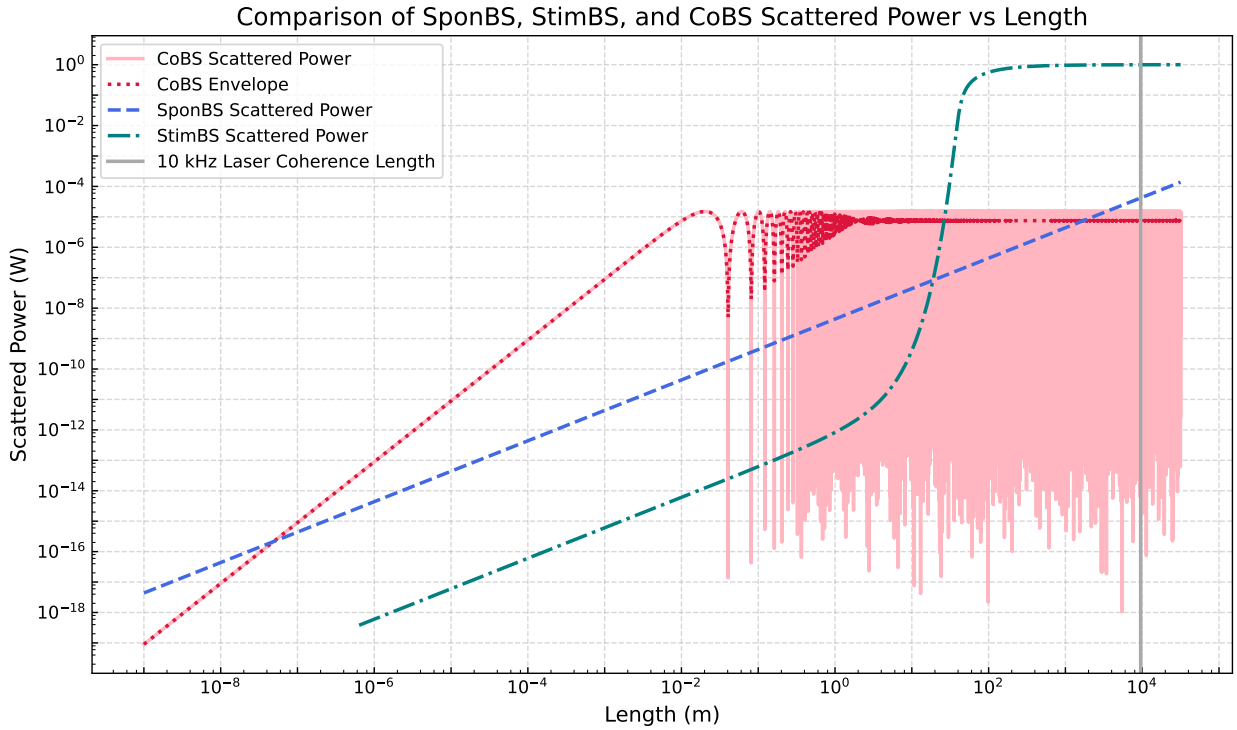


Figure D.2: Comparison of scattered power from a spontaneous Brillouin scattering process and our coherently stimulated Brillouin spectrometer.

the specific sample length being measured such that the scattered power function lies on a local peak of the $sinc^2$ function.

Fig. D.2 shows the advantage that our coherently stimulated Brillouin spectrometer offers compared to the traditional Brillouin processes for the example medium of UHNA3 fiber. For lengths up to about 50 meters and down to as low as 100 nanometers, 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 picoWatts. In contrast, the scattered power for the same system offered by our instrument is on the order of 10s of microWatts, exceeding that of the spontenous process by a factor of a million. This is, of course, the most ideal case for this system, however it can be seen from Fig. D.2 that the coherently stimulated process offers orders of magnitude more scattered power than either traditional process through a wide range of lengths.

D.3 Observance of Fano-Resonant Asymmetries at Small Signals

In *Fano-Resonant Asymmetries at Small Signals* in Sec. **??**) of 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 (Fig. **??**). 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 (CS_2) that we briefly mentioned in Sec. **??** 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 (Sec. **??**) and provide insight into when and why they are most prominent.

D.3.1 Experiment A: Extended 1 cm UHNA3 Fiber Spectra

In 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 sinc^2 dependence on detuning (Fig. **??** 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 Sec. **??** of the main text. As the pump-probe detuning increases, the phase-matching term $\mathrm{sinc}^2(\Delta \mathrm{kL}/2)$ 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.

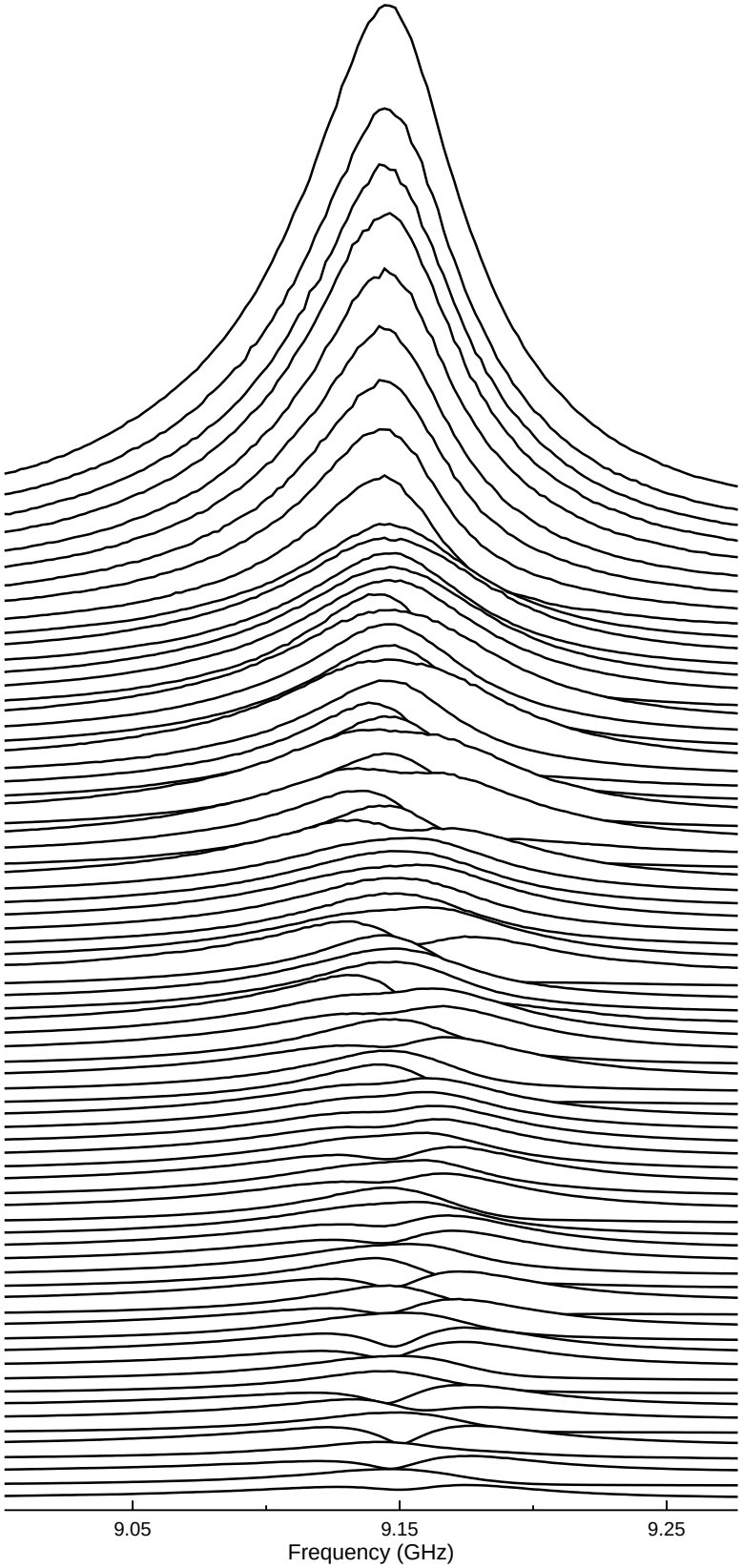


Figure D.3: Stacked Brillouin spectra showing Fano-type line-shape distortions at small signals in a 1 cm UHNA3 fiber. Each trace corresponds to a different pump–probe detuning, revealing the discrete Brillouin resonance (near 9.17 GHz) interfering with a broad continuum background. The resulting asymmetries highlight the characteristic Fano-resonant behavior under low signal conditions.

Figure D.3 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 ($q \to \infty$) at ∼9.17 GHz. By contrast, at detunings between ∼15-20 GHz (where the 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 sinc^2 "lobe,", 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 Sec. **??**. When the Brillouin amplitude significantly exceeds the background, the discrete phonon resonance dominates, resulting in little or no asymmetry ($q \to \pm\infty$). Once the two amplitudes become comparable, Fano interference skews the line shape, shifting the apparent peak frequency slightly and altering the slope on one 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—particularly in small-signal measurements where the Brillouin peak may not tower over the background.

D.3.2 Experiment B: 1 mm CS_2 Spectra and Fano Distortions

We now turn to measurements on a 1 mm-thick cell of CS_2 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 ∼60–70 mW pump, ∼25–30 mW Stokes, and ∼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_2 sample is an order of magnitude shorter (1 mm vs. 1 cm), its phase-matching bandwidth (sinc^2 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^2 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 D.4 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* (Sec. **??**). 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. This lower Brillouin amplitude is owing to the order of magnitude shorter sample length and roughly two orders of magnitude larger effective area in our free-space configuration compared to that of the UHNA3 fiber in Experiment A.

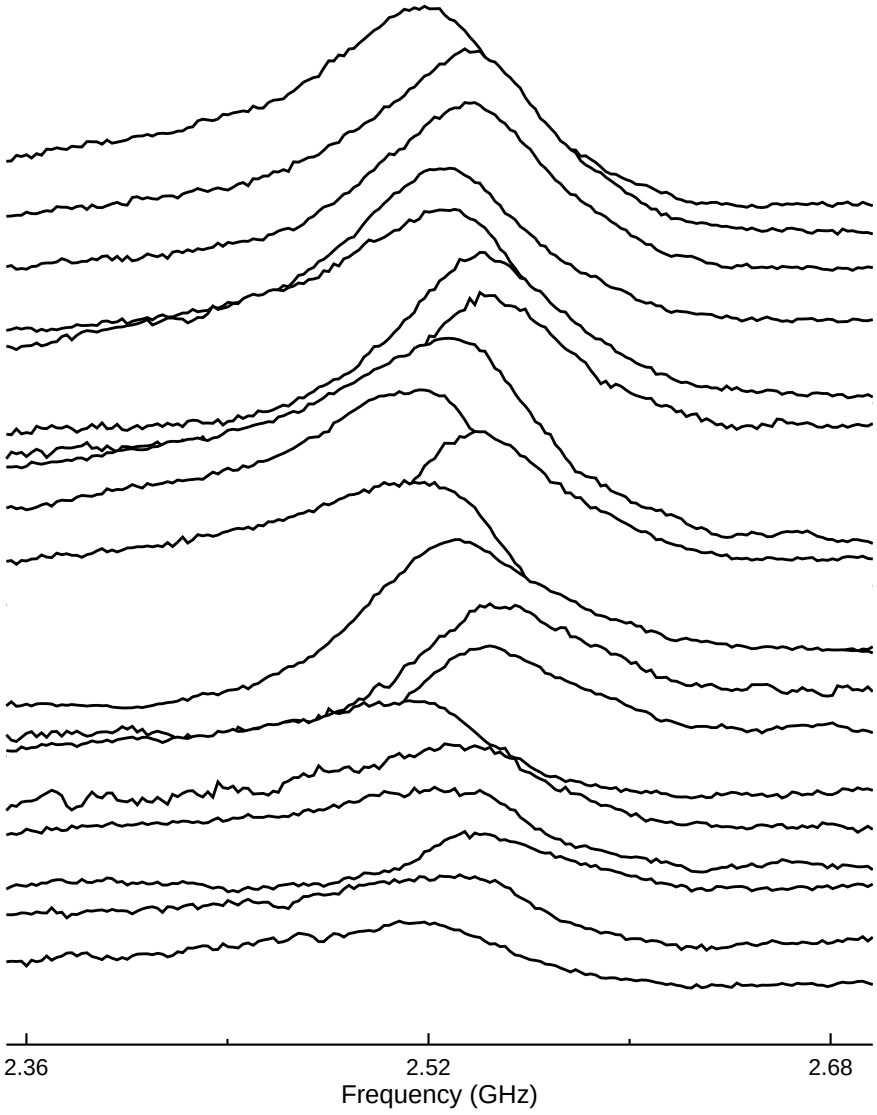
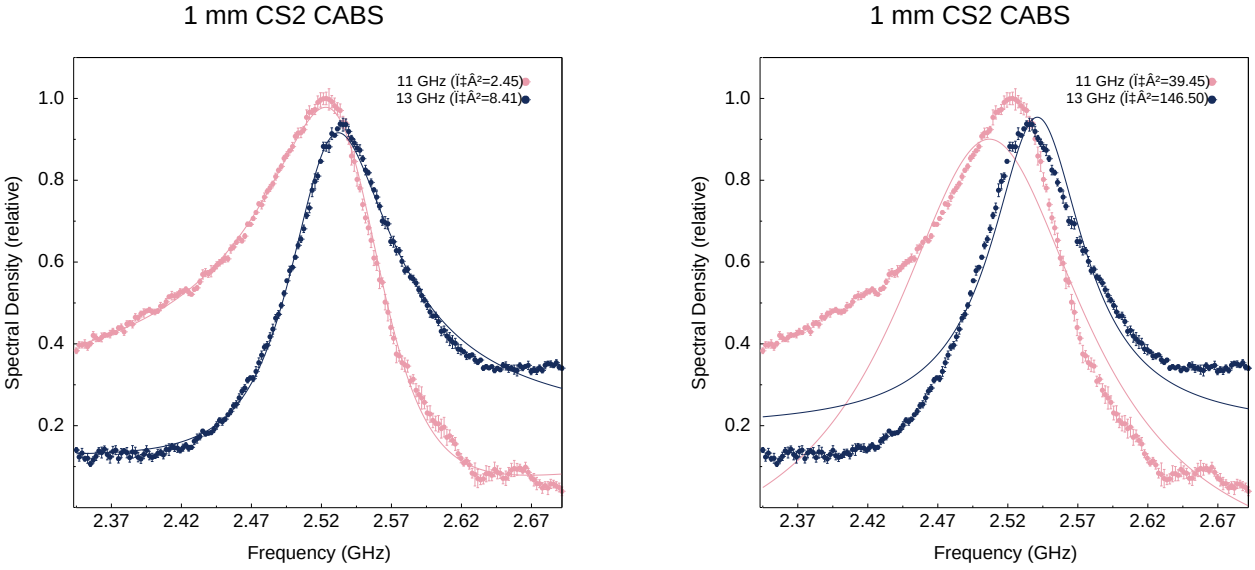


Figure D.4: All measured Brillouin spectra for 1 mm CS_2 at detuning steps of 0.25 GHz from 10 GHz (top spectrum) to 14 GHz (bottom spectrum). Each trace is offset for clarity.

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. Figure D.5 compares 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 in the opposite way, featuring a sharper low-frequency side and a softer high-frequency slope, suggesting $q > 0$. Clearly, the Fano model captures the asymmetric spectral line-shape better than a simple Lorentzian, especially around the half-maximum slopes.

Notably, in the negative-q case (11 GHz), the line's peak amplitude appears slightly higher than what would be inferred from a Lorentzian fit—sometimes referred to as a "peak boost." As discussed in *Fano-Resonant Asymmetries at Small Signals* (Sec. **??**), this can be beneficial for detecting weaker signals, provided the background is not too noisy, and in principle, one could choose a phase relationship that maximizes this constructive interference near resonance. Conversely, a positive-q scenario can suppress or broaden the peak on one side, which may be less desirable for line-shape analysis but could be exploited if one aims to shape the response profile in a particular way.



(a) **(b)**

Figure D.5: Comparison of representative spectra at 11 GHz and 13 GHz, showing the positive vs. negative q asymmetry in 1 mm CS_2.

These 1 mm CS_2 data reinforce the conclusions from the UHNA3 fiber measurements, demonstrating that Fano-type asymmetries arise readily in small-signal regimes across diverse geometries (liquid cell vs. fiber). Moreover, we have explicitly identified positive- and negative-q cases, showing that interference can either boost or clip the Brillouin resonance peak, depending on the sign of q and the relative amplitude of

the background continuum. As in Experiment A, a Fano-fitting procedure is essential to extract accurate linewidths and center frequencies in these small-signal conditions.

To better convey the cyclical evolution of the 75 measured UHNA3 spectra from Experiment A, we have created an animated GIF that steps through each spectrum in ascending pump–probe detuning. We host the GIF (along with the raw data, measurement logs, and plotting scripts) in a public GitHub repository:

`https://github.com/HamletTheHamster/A-Coherently-Stimulated-Brillouin-Spectrometer`

Readers are encouraged to view it for a clear, dynamic perspective on how the line shape transitions between Lorentzian and Fano-distorted forms at different detunings.

D.4 Data

Appendix E

Supplementary Information for Chapter 5: Manuscript III

E.1 Data

References

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