

COHERENTLY STIMULATED BRILLOUIN SPECTROSCOPY

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## **Dedication**



## Preface



# 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

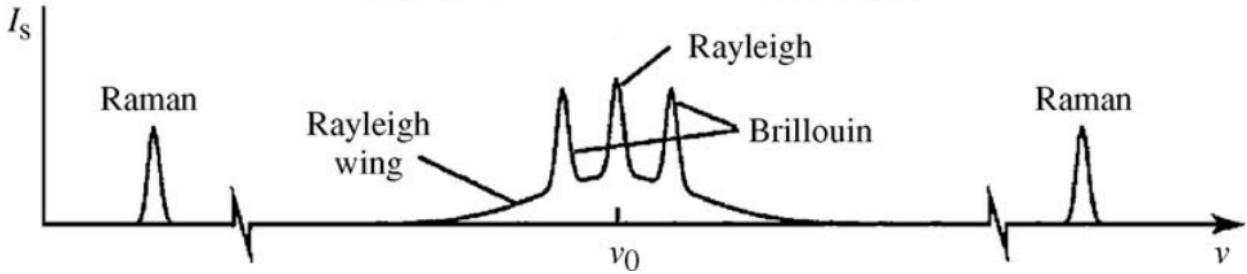
incident light, leading to scattering in possibly a new direction but without a change in wavelength. It is responsible for the blue color of the sky because the efficiency of Rayleigh scattering is inversely proportional to the fourth power of the wavelength ( $\lambda$ ) of the light ( $\frac{1}{\lambda^4}$ ) and so shorter (blue) wavelengths are scattered much more than longer (red) wavelengths by the molecules in the atmosphere.<sup>1</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.

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.<sup>2</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>3</sup>, just one year after the first demonstration of the laser by American physicist Theodor Maiman.<sup>4</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.

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  $\delta z$ , containing a volume element  $\delta V_1$ . For any given incident wavelength  $\lambda$  and any non-zero scattering angle  $\theta$  at volume  $\delta V_1$ , there exists a corresponding volume element  $\delta V_2$ , located a distance  $\frac{\lambda}{2\sin\theta}$  apart, which scatters light at the same angle  $\theta$ . The scattered waves from  $\delta V_1$  and  $\delta V_2$  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.<sup>2</sup> 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.

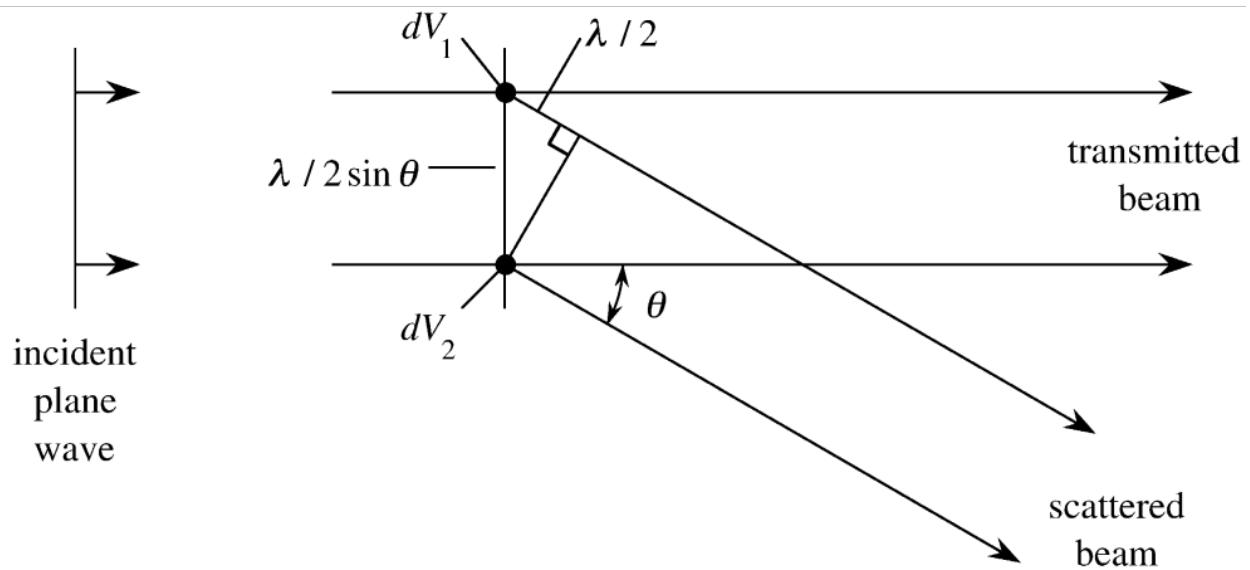


Figure 1.2

## 1.2 Spontaneous Brillouin Scattering

## 1.3 Stimulated Brillouin Scattering

## 1.4 Phase-matching

## 1.5 Brillouin Gain of Materials

## 1.6 Raman Scattering

## 1.7 Raman-like Brillouin Modes



## **Chapter 2**

### **Foundational Experimental Techniques and Instrumentation**

#### **2.1 Photonic Experimental Techniques**

##### **2.1.1 Control of Light in Photonic Systems**

##### **2.1.2 Photonic Devices and Diagrams**

##### **2.1.3 Selection and Isolation of Signals**

##### **2.1.4 Heterodyne Detection and the Local Oscillator**

##### **2.1.5 Optical Loss in a Photonic Systems**

##### **2.1.6 Free Space Optics and Beam Alignment**

##### **2.1.7 Specialized Optical Fibers**

#### **2.2 Optical Instrumentation**

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#### **2.4 Noise and Background Handling**

#### **2.5 Custom Software**

##### **2.5.1 Description of Python Script for CABS Data Collection**

##### **2.5.2 Description of Plotting Data in Go Program**





## Chapter 3

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

### 3.1 Abstract

### 3.2 Optomechanical Cooling and Heating

### 3.3 Cooling Platform: $CS_2$ -Liquid Core Optical Fiber

#### 3.3.1 Optomechanical Properties

#### 3.3.2 Fabrication

#### 3.3.3 Fabrication Iterative Refinement

### 3.4 Intention of the Pump-Probe Experiment

### 3.5 Experimental Setup

#### 3.5.1 Main Experiment

#### 3.5.2 Pump-Probe Experiment

### 3.6 Results

#### 3.6.1 Main Experiment Results

#### 3.6.2 Pump-Probe Experiment Results

### 3.7 Discussion

#### 3.7.1 Application to Ground State Cooling

#### 3.7.2 Standardized Cooling Metric

#### 3.7.3 Tapered chalcogenide Photonic Crystal Fiber: Max Plank Results

## Chapter 4

### Manuscript II: A coherently stimulated phonon spectrometer

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#### 4.1 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. I validated our instrument's sensitivity with 1 cm of 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.

#### 4.2 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>2</sup>. However, the

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weak signal inherent to spontaneous Brillouin scattering often demands long acquisition times and limits spatial resolution, posing challenges for high-resolution material characterization.

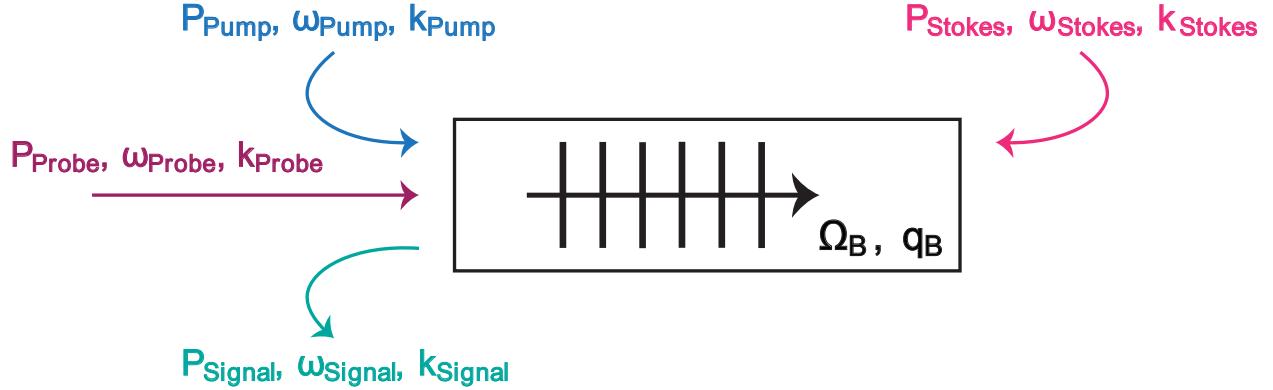
Stimulated Brillouin scattering (SBS) uses intense optical fields to amplify the acoustic wave through a nonlinear optical process<sup>5</sup>. In SBS, a strong pump laser interacts with a counter-propagating Stokes wave in the medium, generating acoustic phonons via electrostriction. As the phonon population 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>6–15</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>16–18</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>19–24</sup>. Techniques based on optical cavities increase the effective interaction length, but require precise alignment and are sensitive to environmental fluctuations<sup>25</sup>. Forward Brillouin scattering methods, such as those demonstrated by Kittlaus et al.<sup>26</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>27</sup>.

Here, I 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. I demonstrate the capabilities of our instrument by measuring Brillouin scattering in 1 cm of ultrahigh numerical aperture (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 spectrometer opens new avenues for nanometer-scale Brillouin spectroscopy and advances the characterization and design of nano-acousto-optic devices. It



**Figure 4.1:** Illustration of 4-Wave Brillouin Scattering.

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.

### 4.3 Theoretical Framework

#### 4.3.1 Coherently stimulated four-wave Brillouin scattering

Traditional SBS, illustrated by the schematic in Figure 4.1(a) for the Stokes process, 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 4.1(b) illustrates coherently stimulated four-wave Brillouin scattering for the Stokes process. I 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, I 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, I derive the coupled-wave equations for the four-wave mixing process in Appendix D.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,

$$P_{\text{Sig}} = \frac{1}{4} (G_B L)^2 P_P P_S P_{Pr} \text{sinc}^2 \left( \frac{\Delta k L}{2} \right), \quad (4.1)$$

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

$$G_B = \frac{g_0}{A_{\text{eff}}} \frac{\left(\frac{\Gamma_B}{2}\right)^2}{(\Omega - \Omega_B)^2 + \left(\frac{\Gamma_B}{2}\right)^2}, \quad (4.2)$$

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

$$g_0 = \frac{\gamma_e^2 \omega^2}{n v c^3 \rho_0 \Gamma_B}. \quad (4.3)$$

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 4.2,  $\Omega_B$  is the resonant Brillouin frequency of the material,  $A_{\text{Eff}}$  is the effective area of 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.

### 4.3.2 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>28</sup> This can be seen by Equation 4.1, 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

$$\Delta k = \frac{4\pi n \Delta \lambda}{\lambda_{\text{Pump}} \lambda_{\text{Probe}}} \approx \frac{4\pi n \Delta \lambda}{\lambda_{\text{Pump}}^2}. \quad (4.4)$$

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,

$$\Phi \equiv \text{sinc}^2 \left( \frac{2\pi n \Delta \lambda L}{\lambda_{\text{Pump}}^2} \right). \quad (4.5)$$

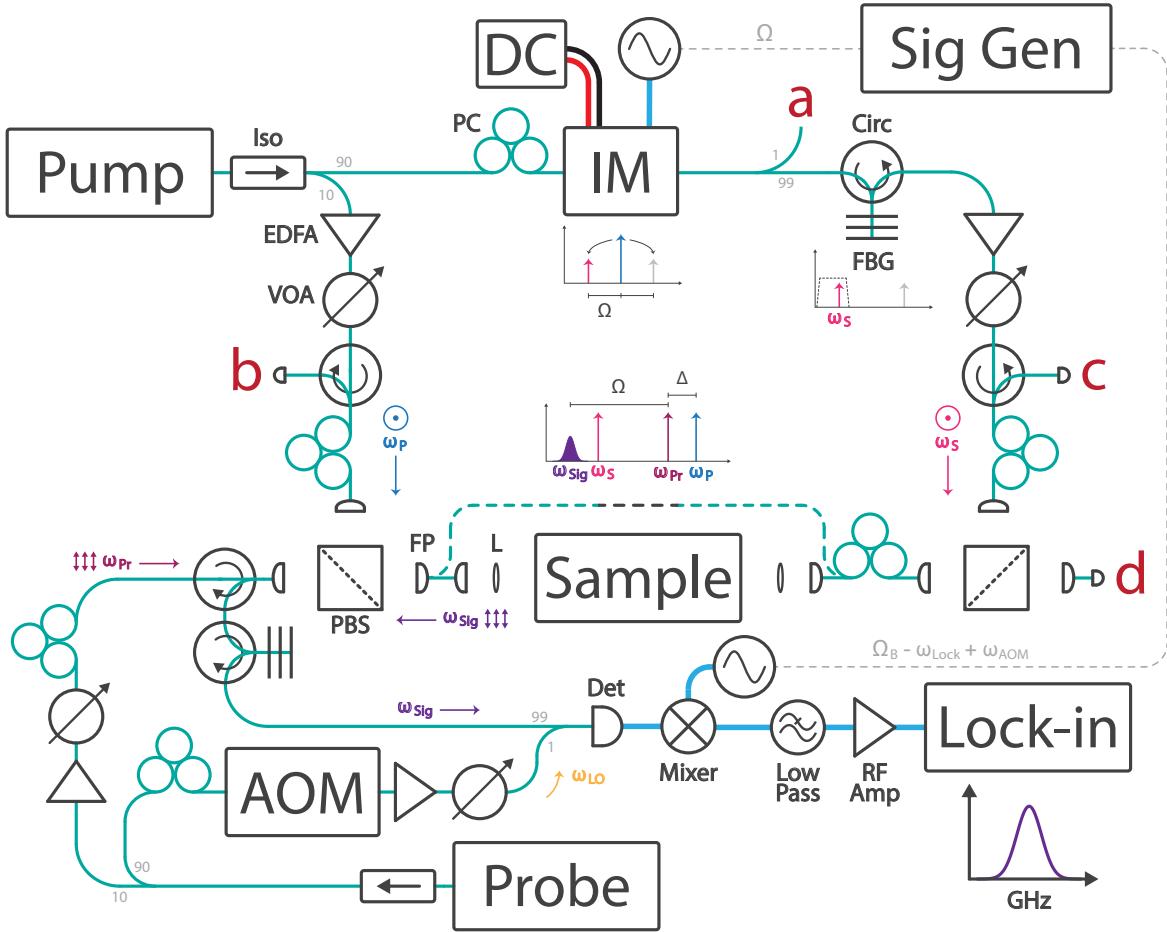
Using this expression for  $\Phi$ , I see that for an effective length of 1 m, a wavelength mismatch of only 0.6 pm from a typical wavelength of 1.55  $\mu\text{m}$  pump light in UHNA3 fiber drops the scattered power to half of the maximum. However, for shorter effective lengths the wavevector mismatch 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 4.5 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 4.1, 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.

## 4.4 Methods

### 4.4.1 Instrument design

Figure 4.2 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 (EDFA) and passed through a variable optical attenuator (VOA) for power control. The output is



**Figure 4.2:** Design schematic of a coherently stimulated phonon spectrometer. 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 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 an 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, AOM-shifted local oscillator (LO) derived from the probe laser and directed to a photodiode for detection. The resulting RF signal is mixed with an 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.

then polarization-controlled to reflect at a polarizing beam splitter (PBS) for injection into the sample. For Stokes synthesis, an AC signal ( $\Omega$ ) is supplied to an intensity modulator (IM) 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 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 (LO). 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 (AOM) ( $\omega_{\text{LO}} = \omega_{\text{Probe}} + \omega_{\text{AOM}}$ ) 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 (RF) 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.

#### 4.4.2 Experimental Techniques

We optimized the signal-to-noise ratio (SNR) of our 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 local oscillator (LO). To combine the backscattered signal and LO with minimal loss, I 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. I find that running both the pump and probe lasers in “whisper” mode (as opposed to “dither”) significantly enhances the measured SNR.

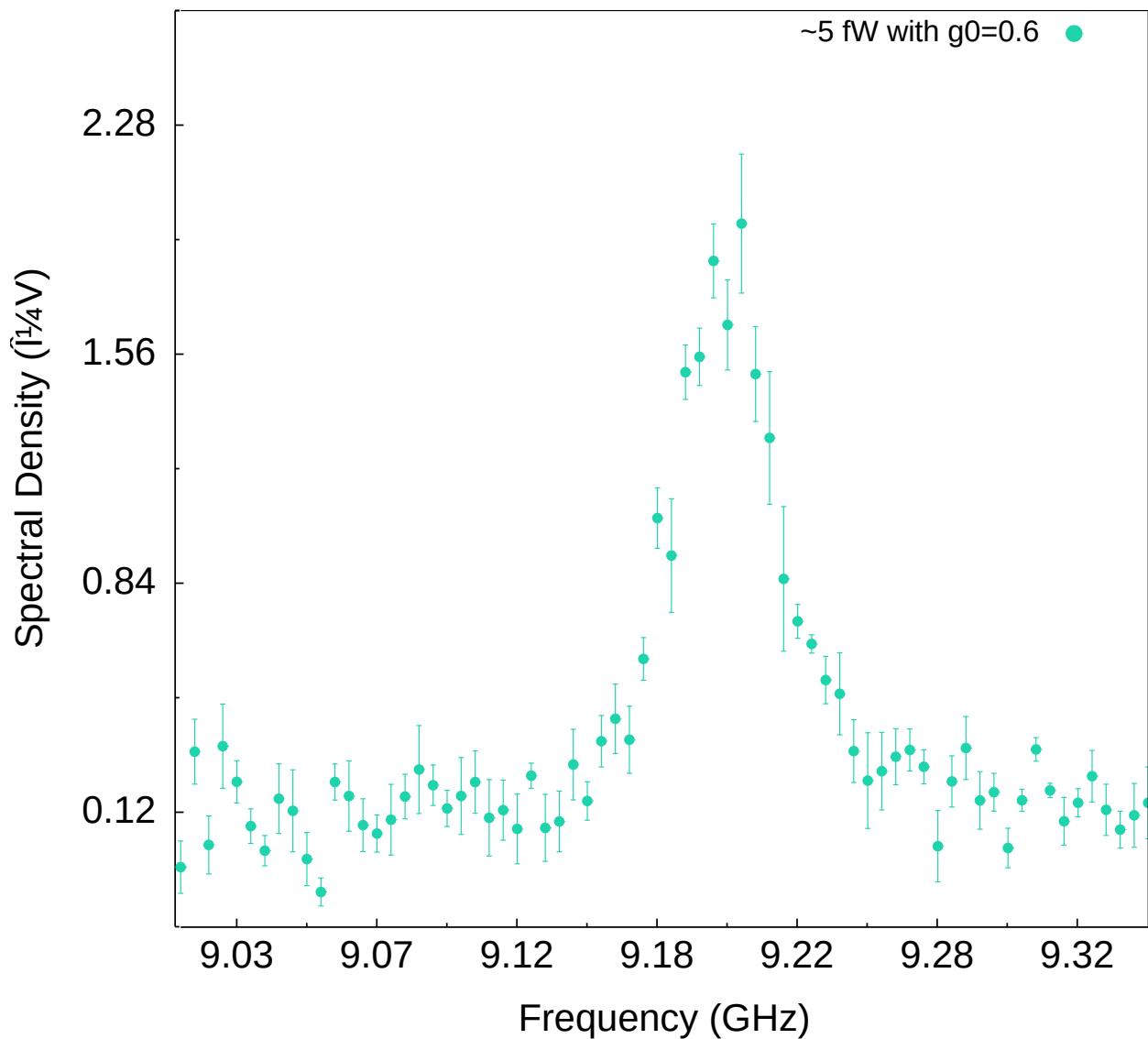
We use a Zurich Instruments HF2LI 50 MHz lock-in amplifier whose demodulator settings are carefully tuned to maximize SNR. A 10 MHz reference clock from the signal generator is fed into the lock-in to synchronize timing. The input-signal range, which sets the analog input amplifier’s gain, should exceed the measured signal (including any 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. I 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, I 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, I observe less than 100 Hz of drift in the detected signal frequency, so I typically set a 100 Hz low-pass bandwidth for multi-hour measurements. For shorter scans ( $< 15$  min), I 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.

## 4.5 Results

### 4.5.1 Instrument sensitivity

## 1 cm UHNA3 CABS



**Figure 4.3:**  $\sim 5$  fW sensitivity measurement

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 4.1, the sensitivity of the instrument is the minimum scattered power,  $P_{Sig}$ , to produce a statistically significant measurement. To determine this, I target a specific length,  $L$ , of a sample of known effective Brillouin gain,  $G_B$ . I 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, I gradually reduce the optical powers until the sensitivity floor is reached.

To serve as our sensitivity testbed, I prepared 1 cm of Nufern's UHNA3 fiber, a well-studied fiber with known effective Brillouin gain<sup>29</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 subsequent reduction in optical powers. First, it offers a Brillouin shift that is spectrally far from that of the single-mode fiber (SMF28) 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>29</sup>. This gain factor is larger than that of SMF28 by an order of magnitude<sup>30</sup>.

Figure 4.3 presents a measurement in which the instrument's sensitivity reaches  $P_{Sig} = 5 \text{ fW}$ . 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, I 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.

$G_B$ ( $\text{W}^{-1} \text{ m}^{-1}$ )	$L$ (m)	$P_P$ ( $\mu\text{W}$ )	$P_S$ ( $\mu\text{W}$ )	$P_{Pr}$ ( $\mu\text{W}$ )	$\Delta\lambda$ (pm)
0.6	0.01	506	504	2.01	20

**Table 4.1:** Measurement parameters for sensitivity measurement and calculation.

### 4.5.2 Measurements

We demonstrate the capabilities of the instrument on two common sample classes: fiber and bulk material. For a fiber sample I again choose UHNA3 for its higher nonlinear response and excellent optical and acoustic

guidance. In contrast to the sensitivity measurements, I 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. I target the same 1 cm segment of UHNA3 fiber as was used for determining sensitivity.

## 1 cm UHNA3 CABS

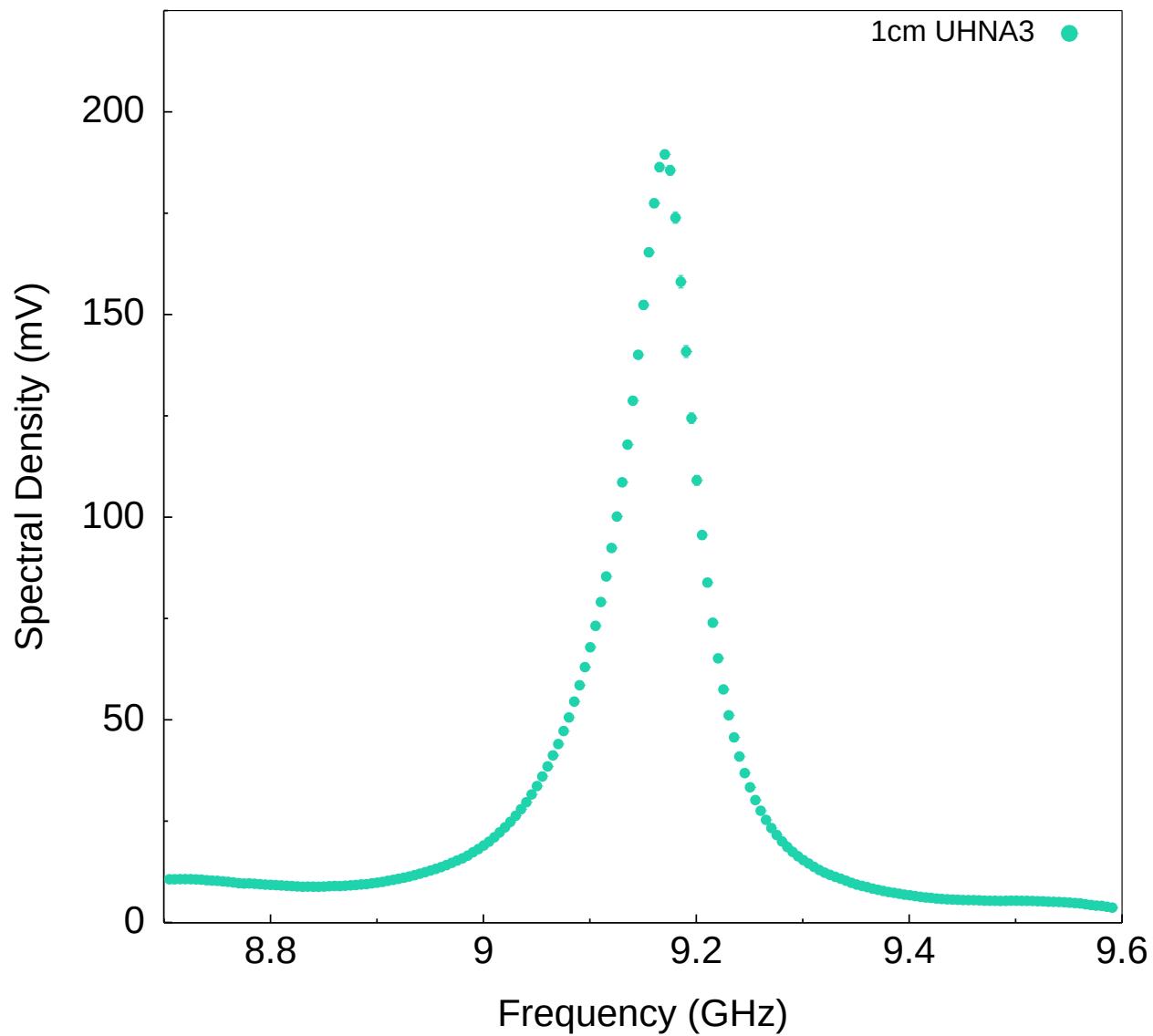
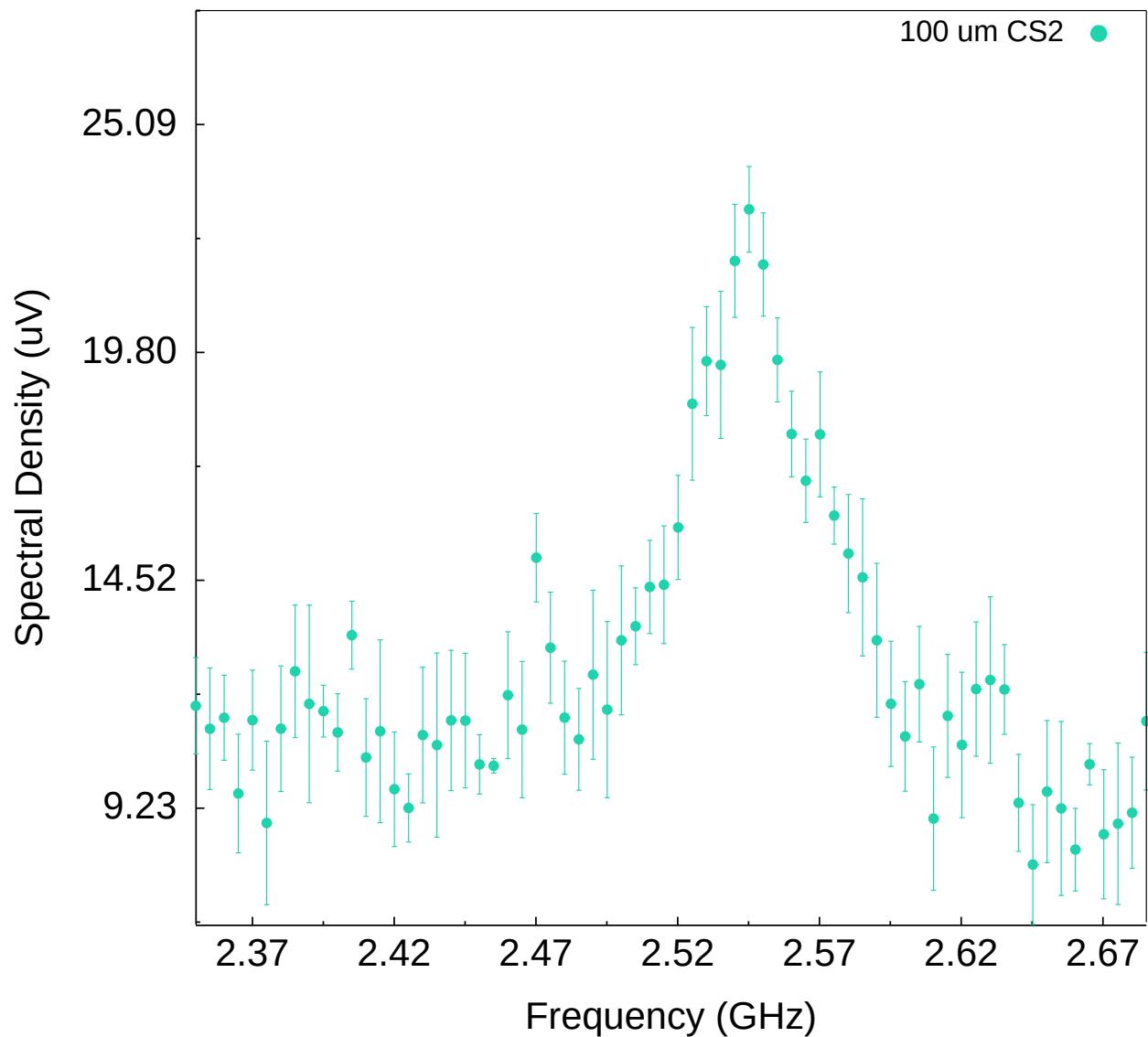


Figure 4.4: 1 cm UHNA3

Figure 4.4 shows the spectral profile captured for 1 cm of UHNA3 fiber, revealing the expected lorentzian profile consistent with Equation 4.2. 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 FWHM 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>29</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. I 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 pass-band 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. I placed the 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.

## 100 $\frac{1}{4}$ m CS2 CABS



For a free-space bulk example I target liquid CS<sub>2</sub> for its exceptionally high Brillouin gain factor of 1.5 mGW<sup>-1</sup>.<sup>2</sup> Figure 4.5 reveals the Brillouin signal of bulk CS<sub>2</sub> liquid contained in a 100 μm 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 CS<sub>2</sub>. 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 CS<sub>2</sub> 2.55 GHz shift. I 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 4.5, this corresponds to a Stokes and probe filter placement of 1548.844 nm and 1548.934 nm, respectively.

#### 4.5.3 Phase Matching Bandwidth

To characterize the phase matching tolerance of the instrument for a given length of sample, I performed an additional experiment whereby I 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 4.1, 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 4.1, I would expect the peak amplitudes of these measurements to trace out a sinc<sup>2</sup> profile, also given by Equation 4.5. Figure 4.6 shows the results of this experiment, in which 75 measurements performed between 5 GHz and 42 GHz pump–probe frequency separation at 0.5 GHz intervals. I found peak amplitudes by fitting each spectra with a Fano profile (see the proceeding *Fano-Resonant Asymmetries at Small Signals* subsection) and are represented by a point in Figure 4.6. The theoretical sinc<sup>2</sup> function for the parameters used in the experiment is shown on the plot with a solid red line.

## 1 cm UHNA3 Phase-Matching Bandwidth

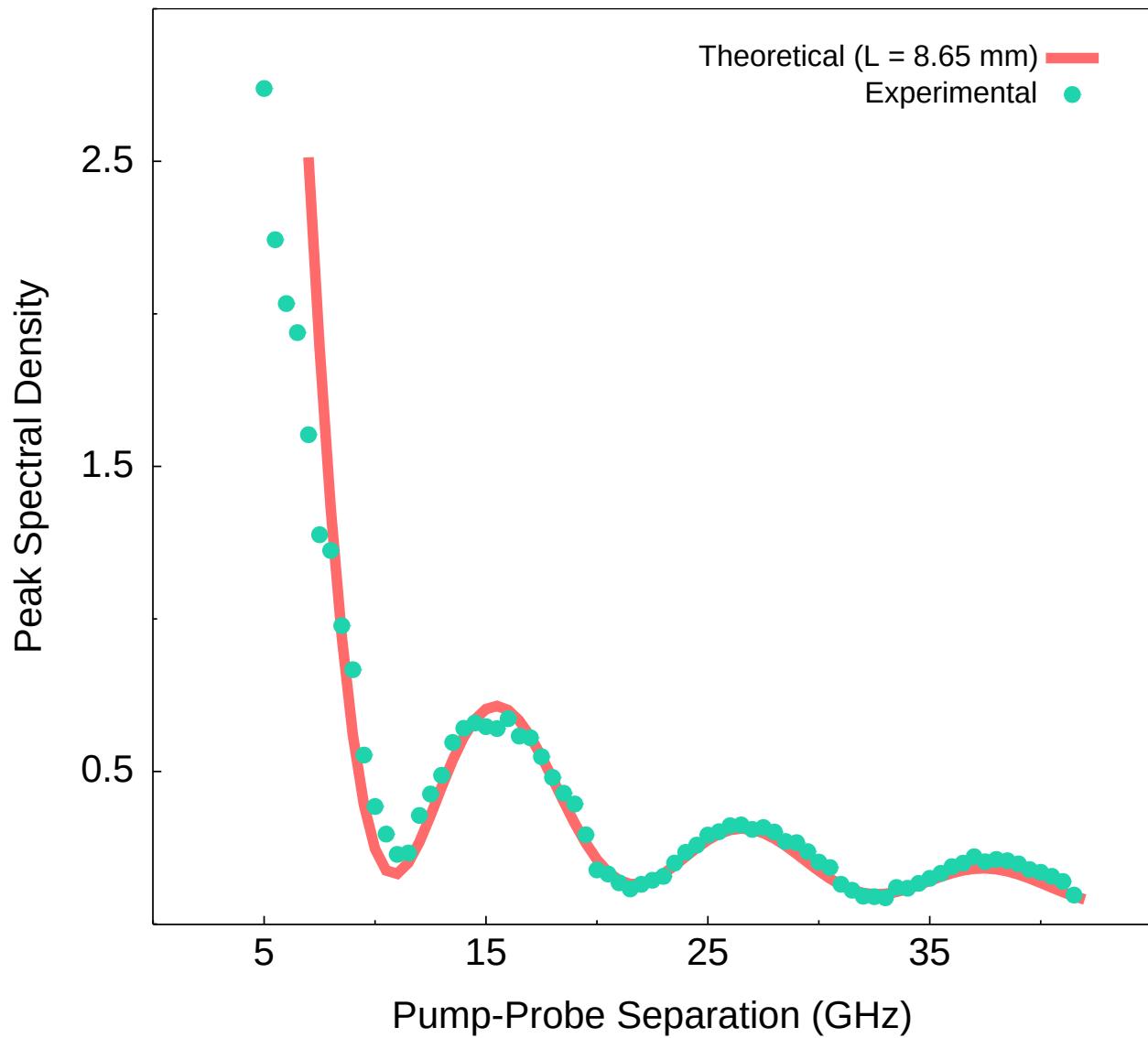


Figure 4.6: Phase-matching sinc<sup>2</sup> func

#### 4.5.4 Fano-Resonant Asymmetries at Small Signals

Under certain conditions where the resonant Brillouin amplitude approaches the background continuum level, I observe an asymmetric, Fano-type line-shape<sup>31–34</sup>. These Fano distortions can shift the apparent peak frequency, complicate simple Lorentzian fitting, and affect the extracted linewidth in small-signal measurements.<sup>35</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>31</sup>,

$$I(\omega) \propto \frac{(q + \epsilon)^2}{1 + \epsilon^2}, \quad (4.6)$$

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

$$q = \frac{(\text{resonant amplitude})}{(\text{background amplitude})} \cot(\Delta\phi), \quad (4.7)$$

where  $\Delta\phi$  is the phase difference between the oscillation driven by the discrete resonant mode and that of the background continuum<sup>32;36;37</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>32</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 4.6. If they are exactly in phase ( $\Delta\phi = 0 \text{ or } \pi$ , 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 4.5) and 1 cm UHNA3 fiber at low power (Figure 4.3), 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 4.6 can become finite rather than  $\pm\infty$  in the limit that the background is negligible, and Fano interference arises. To explore this further, I 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 D.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 D.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 D.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 signal-to-noise ratio 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, I can achieve this without sacrificing independent control of the pump–probe detuning: by simultaneously shifting the pump laser in step with the probe, I 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 4.6), 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 D.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, I more accurately capture the asymmetric peak and its shifted center frequency.

## 4.6 Conclusion

In conclusion, I 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 5**

### **Manuscript III: Brillouin-induced Raman modes**

#### **5.1 Abstract**

#### **5.2 Introduction**

#### **5.3 Methods**

#### **5.4 Results**

#### **5.5 Discussion**



## **Chapter 6**

### **Discussion & Future Work**



## 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

Table

## Appendix D

### Supplementary Information for Chapter 4: Manuscript II

#### D.1 Coupled-Wave Equations

Here I derive the coupled wave equations that describe coherent stimulated Brillouin scattering involving a pump, Stokes, probe, and backscattered optical field given respectively by

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

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

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

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

and a common acoustic field given by

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

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

### D.1.1 Acoustic Field

As in the case of SBS<sup>2</sup>, I start by assuming that the material obeys the acoustic wave equation,

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

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

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

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 D.6 is the divergence of the electrostrictive force:

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

which yields, after applying the slowly varying amplitude approximation,

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

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 and the acoustic field (Equation D.5) into Equation D.6, and assuming a slowly varying acoustic amplitude, I find

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

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

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

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

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

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 D.12 to be dropped. I further assume steady-state conditions such that the time derivative term also vanishes, leaving

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

We thus find the acoustic field amplitude to be

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

### D.1.2 Optical Fields

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

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

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

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

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

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

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

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

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

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

$$\frac{\partial A_P}{\partial z} + \frac{n}{c} \frac{\partial A_P}{\partial t} = \frac{i\omega_P \gamma_e}{2nc\rho_0} \rho A_2, \quad (\text{D.21})$$

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

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

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

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

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

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

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

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

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

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

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

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

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

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

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

giving

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

and in fact on resonance the expression reduces to

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

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

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

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

$$g_0 = \frac{\gamma_e^2\omega^2}{nvc^3\rho_0\Gamma_B}, \quad (\text{D.37})$$

reduces this expression to

$$A_{Sig} = -\epsilon_0ncg_0 A_PA_S^*A_{Pr} \frac{e^{-i\Delta k L} - 1}{-i\Delta k}. \quad (\text{D.38})$$

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

$$I_i = 2n\epsilon_0c|A_i|^2, \quad i = 1, 2, 3, \dots \quad (\text{D.39})$$

which produces for the signal intensity

$$I_{Sig} = 2\epsilon_0nc(\epsilon_0ncg_0)^2|A_P|^2|A_S^*|^2|A_{Pr}|^2 \left| \frac{e^{-e\Delta k L} - 1}{-i\Delta k} \right|^2 = 2\epsilon_0^3\epsilon_0 n^3 c^3 g_0^2 \frac{I_P}{2\epsilon_0 nc} \frac{I_S}{2\epsilon_0 nc} \frac{I_{Pr}}{2\epsilon_0 nc} \left| \frac{e^{-e\Delta k L} - 1}{-i\Delta k} \right|^2. \quad (\text{D.40})$$

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

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

giving as a final expression for backscattered signal intensity,

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

To find the power of the backscattered signal, I would integrate this intensity over the effective area. For a uniform area  $A_{eff}$ , this gives

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

or,

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

where,

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

We can also see that off resonance, the  $\Omega$  term from Equation D.34 goes to a lorentzian form after taking the squared modulus for intensity.

## 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 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 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 quadratic dependence on length in the overall scattered power, as seen in Equation 4.1. 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 I 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>2</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  $W^{-1}m^{-1}$  ( $G_B = \frac{g}{A_{eff}}$ ),  $P_P$  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<sup>38</sup> as

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

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. Rearranging this Equation and converting to effective Brillouin gain,  $G_B$ , and power by applying the effective area, I arrive at the scattered power of the Stokes spontaneous Brillouin scattering process,

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

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

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

to be a good approximation. I thus find that

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

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

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

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>39</sup> for the complementary forward scattering process. 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 I 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>38</sup> whereas for typical lengths of single mode fiber it can be lower<sup>9</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>38</sup>

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

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, I solve for the scattered power of the Stokes field,

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

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 D.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>2</sup> To account for pump depletion, I 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

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

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

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

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 D.1, along with the analytical solutions derived previously for the low-gain (spontaneous) regime and our coherently stimulated Brillouin spectrometer given by Equation 4.1. System parameters used to generate the plot for each of the three processes are provided in Tables D.1 and D.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.

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

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

**Table D.1:** 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.

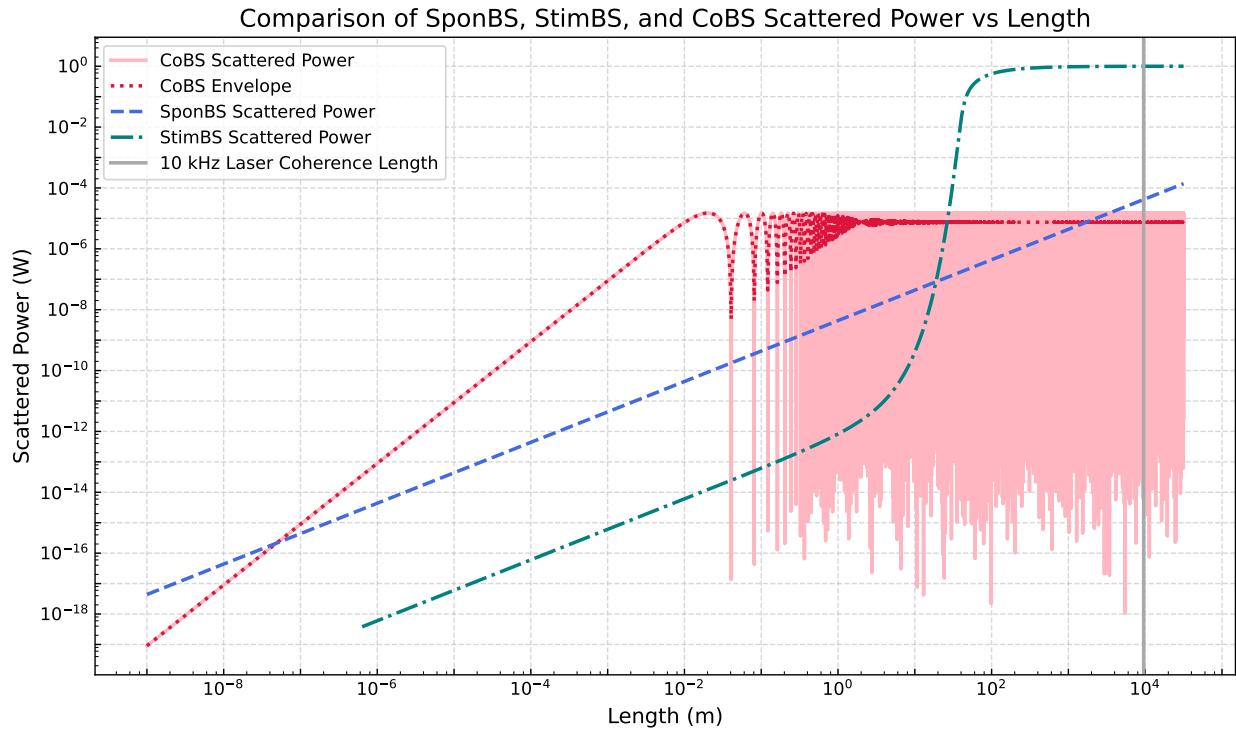
At lengths beyond a centimeter, the phase-matching relaxation of the coherently stimulated process begins

### Spontaneous and Stimulated Scattering Process Model System Parameters

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

**Table D.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.

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 Equation 4.5. 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, I 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  $sinc^2$  function.



**Figure D.1:** Comparison of scattered power from a spontaneous Brillouin scattering process and our coherently stimulated Brillouin spectrometer.

Figure D.1 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 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 D.1 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

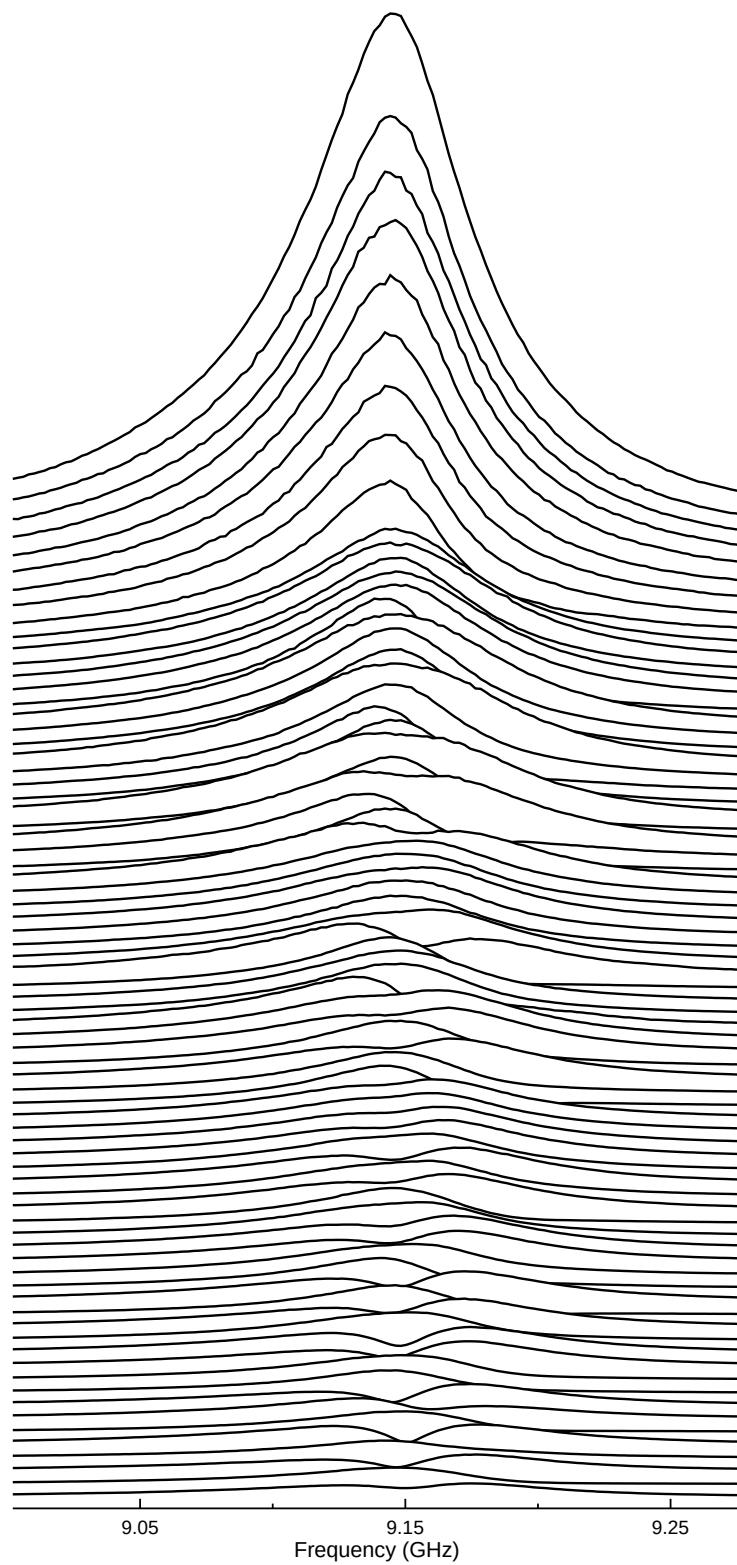
In *Fano-Resonant Asymmetries at Small Signals* (Section 4.5.4 in the main text), I discussed how Fano-type interference can distort Brillouin line shapes in situations where the resonant Brillouin amplitude becomes comparable to the background continuum. I focus here on two experiments (A and B) that 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 4.6). Here I 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 I briefly mentioned in Section 4.5.4 but did not detail. This experiment was performed specifically to further probe the unexpected Fano-like distortions observed in Experiment A. In each case, I 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 4.5.4) and provide insight into when and why they are most prominent.

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

In Section 4.5 of the main text, I 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, I reported only the resulting peak amplitudes, showing how they follow a  $\text{sinc}^2$  dependence on detuning (Figure 4.6 in the main text). However, each measurement in that scan also yields a full Brillouin spectrum—75 in total. Here, I 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). I used the same setup and procedure described in *Phase Matching Characterization* in Section 4.5 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 D.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 4.5. 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 D.2:** All measured Brillouin spectra for 1 cm UHNA3 at 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.

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

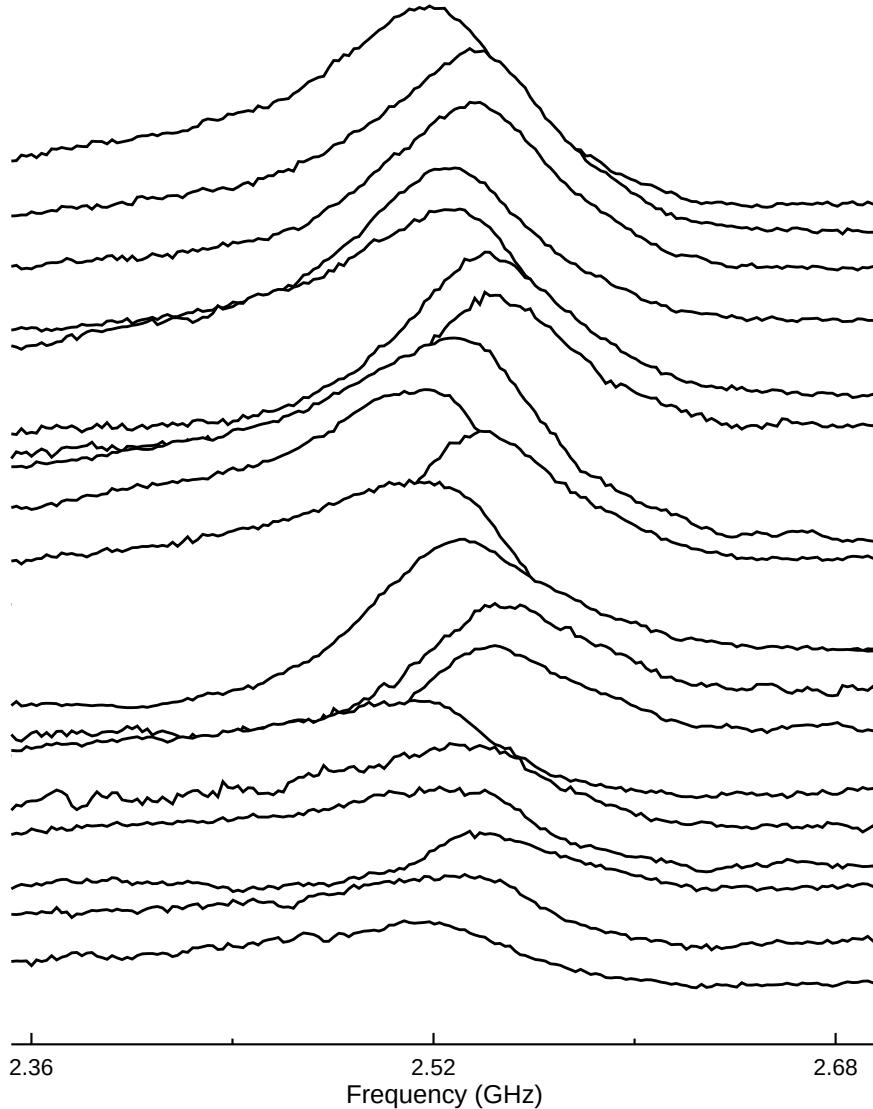
We now turn to measurements on a 1 mm-thick cell of CS<sub>2</sub> in a free-space geometry, complementing the 1 cm UHNA3 fiber results (Experiment A). Both experiments used comparable sub-Watt optical powers (on the order of  $\sim$ 60–70 mW pump,  $\sim$ 25–30 mW Stokes, and  $\sim$ 40–50 mW probe). 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 D.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 4.5.4). 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. 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 4.1 and 4.2 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-

robicates 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 4.6). 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.



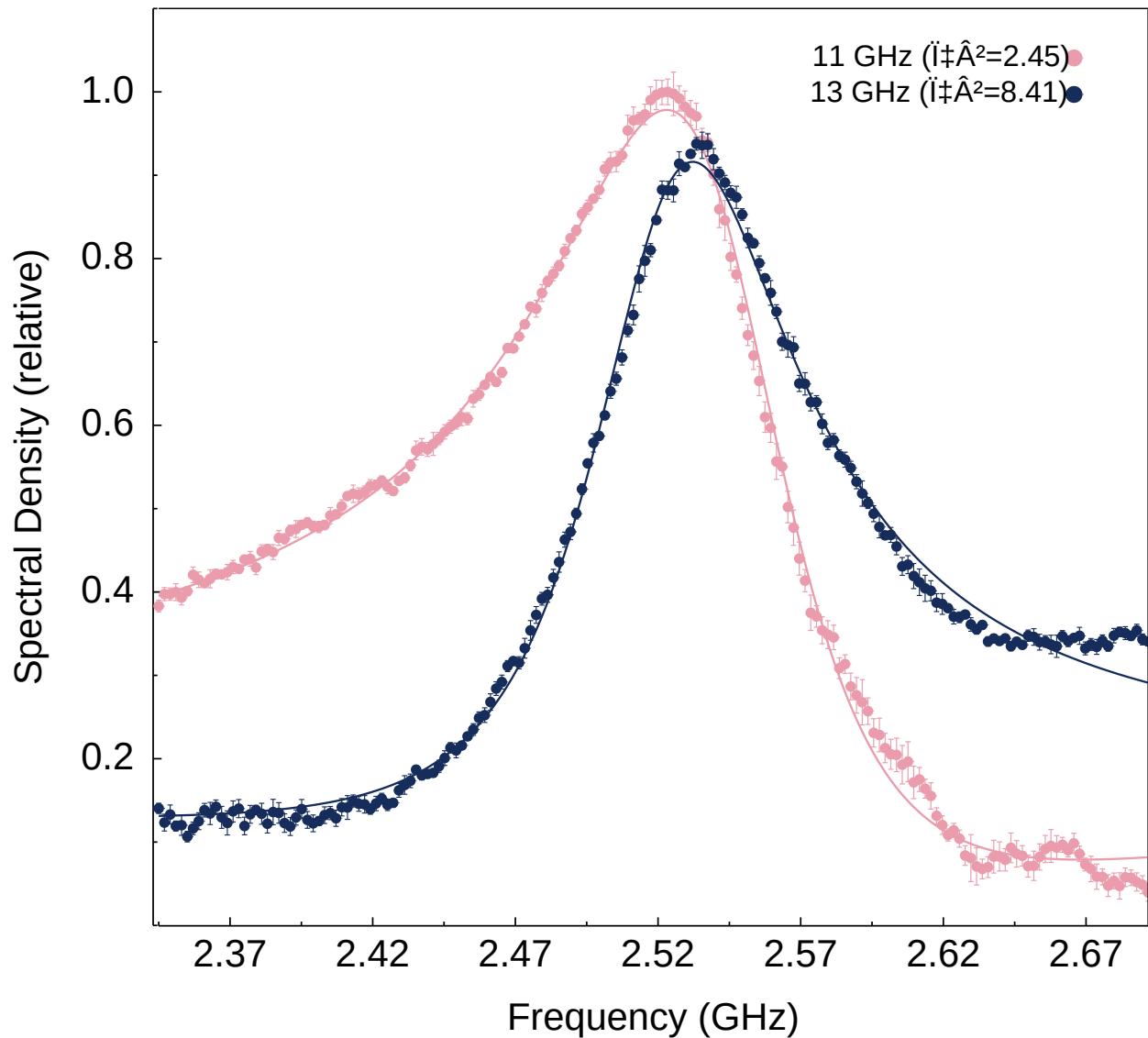
**Figure D.3:** All measured Brillouin spectra for 1 mm  $\text{CS}_2$  at detuning steps of 0.25 GHz from 10 GHz (top spectrum) to 14 GHz (bottom spectrum). Each trace is offset for clarity.

To illustrate the strong distinction in line-shape of a spectra resulting from a positive vs. negative  $q$  value, I focus on two particular detunings that yielded notably skewed line shapes: 11 GHz and 13 GHz. Figures D.4 and D.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 D.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 D.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.

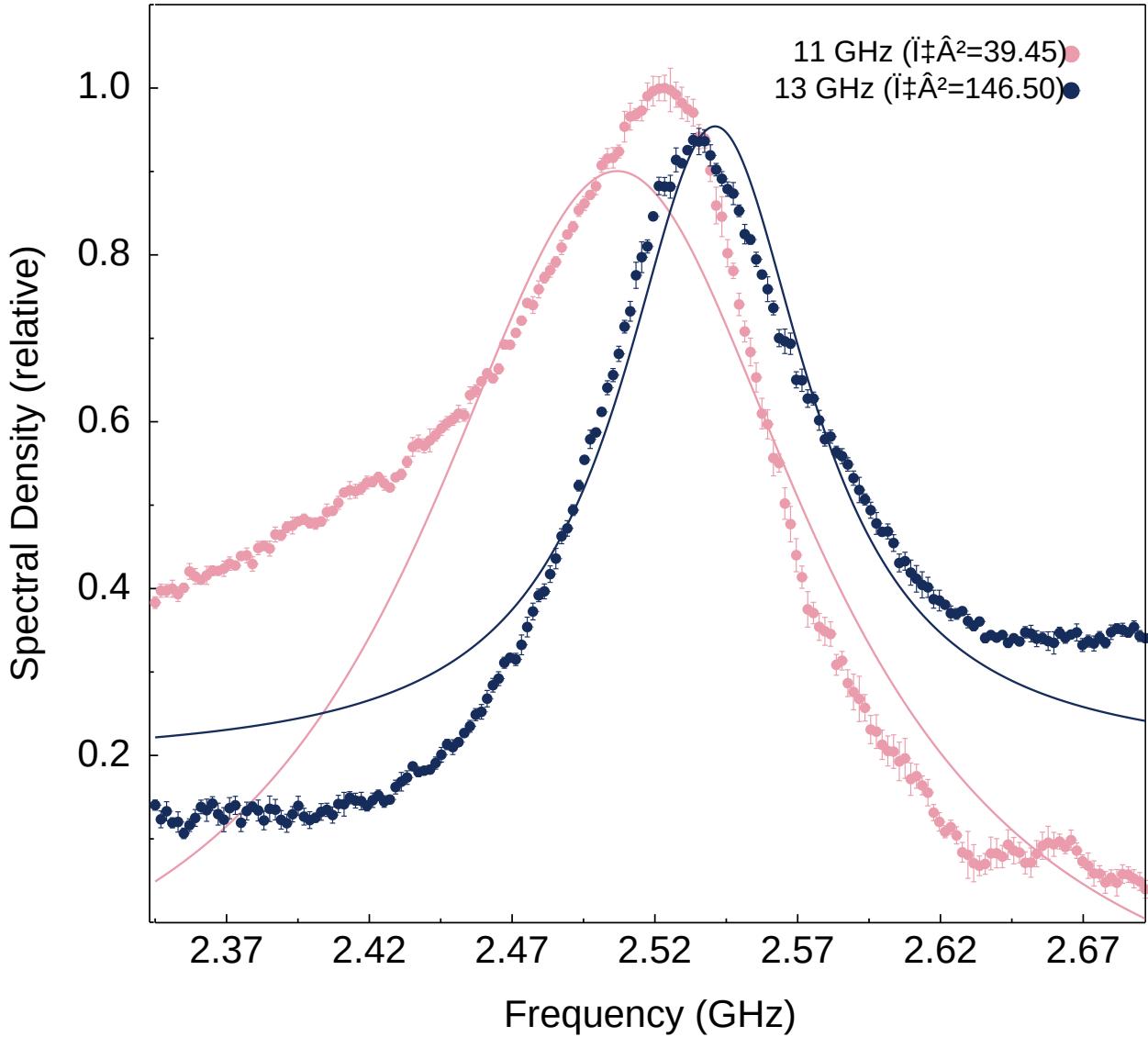
Notably, in the 11 GHz case, our Fano fit reveals a local amplitude slightly higher than what the Lorentzian fit suggests, sometimes referred to as a “peak boost.” In essence, partial *constructive* interference between the discrete Brillouin response and the broad continuum locally raises the amplitude, although it does not imply any net energy gain. As mentioned in *Fano-Resonant Asymmetries at Small Signals* (Section 4.5), 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>36;37;40</sup>

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



**Figure D.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 D.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, I have created an animated GIF that steps through each spectrum in ascending pump–probe detuning. I 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 Equal Contribution of Pump, Stokes, and Probe Optical Fields

Equation 4.1 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.

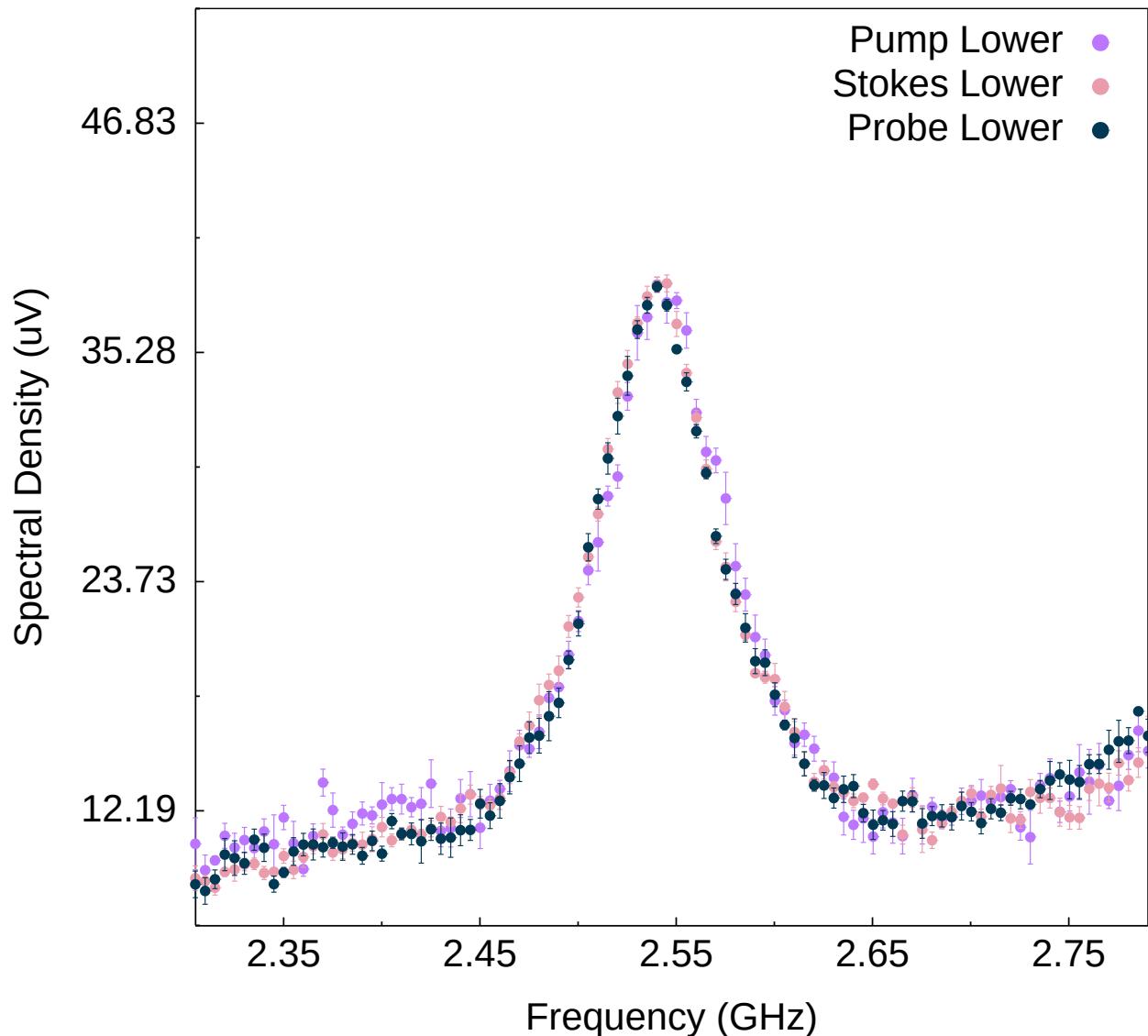
To test this, I conducted a controlled experiment with a 1 mm carbon disulfide ( $CS_2$ ) sample. For each measurement, one of the three source powers (Pump, Stokes, or Probe) was systematically reduced by 75% while holding the others constant and ensuring consistent experimental conditions across trials. Table D.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 D.3:** Power values for each source (Pump, Stokes, Probe) across the three measurements, with the multiplicative total power for each setup.

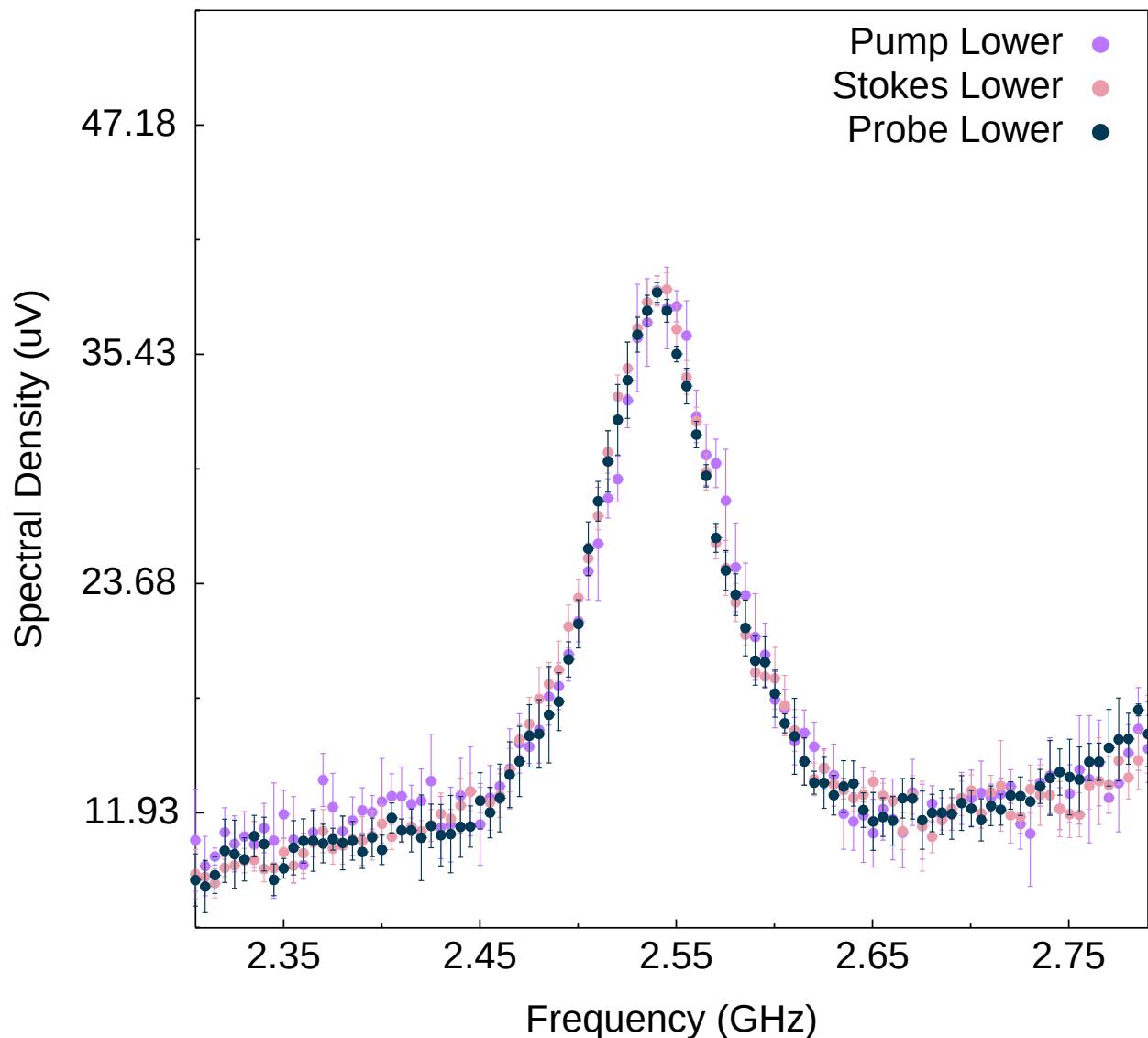
Figure D.6 displays the average results from these three measurements, plotted with error bars representing one standard deviation of the mean. For increased certainty, Figure D.7 presents the same data with error bars extended to two standard deviations, providing additional confidence in the reproducibility 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 4.1 for predicting Signal power across a range of power distributions within practical settings.

## 1 mm CS2 CABS



**Figure D.6:** Signal power contributions with error bars representing one standard deviation of the mean for each measurement.

## 1 mm CS2 CABS



**Figure D.7:** Signal power contributions with error bars extended to two standard deviations of the mean for each measurement.

## D.5 Data



## **Appendix E**

### **Supplementary Information for Chapter 5: Manuscript III**

#### **E.1 Data**



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