SMALL-SCALE COHERENTLY STIMULATED BRILLOUIN SPECTROSCOPY

By Joel N. Johnson

A Dissertation

Submitted in Partial Fulfillment

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Doctor of Philosophy

in Applied Physics and Materials Science

Northern Arizona University

!Month YYYY!

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Dedication

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Preface

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Chapter 1

Introduction

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

Optomechanics comprises a broad range of phenomena involving the interaction of optical and mechanical systems, from basic photothermal absorption to more complex nonlinear processes. Here I offer a brief overview of notable optomechanical phenomena then devote the remainder of this chapter to a more detailed description of the specific interactions that play a role in my research. Photothermal absorption is the process by which light is absorbed by a material, leading to an increase in temperature of the material and consequent changes in the material's dimensions (thermal expansion) or refractive index (thermo-optic effect). This effect has applications in optical switches , actuators , and sensors . Photothermal therapy in medicine is an emerging application of this effect, where light is used to target and heat specific areas, causing localized damage to diseased tissue . This technique becomes especially effective when combined with nanoparticle-enhanced absorption, allowing for dramatically increased absorption in ultra-localized zones within the body.

Light scattering, in its many forms, is also an optomechanical process as it involves the interaction of an optical field with the fluctuation, motion, or vibration of matter. Rayleigh scattering, perhaps the most well-known example, is the elastic scattering of light by particles much smaller than the wavelength of the incident light, leading to scattering in possibly a new direction but without a change in wavelength. It is responsible for the blue color of the sky because the efficiency of Rayleigh scattering is inversely proportional to the fourth power of the wavelength (λ) of the light ($\frac{1}{\lambda^4}$) and so shorter (blue) wavelengths are scattered much more than longer (red) wavelengths by the molecules in the atmosphere.

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

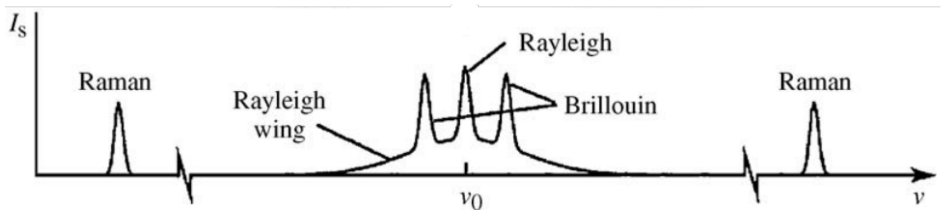

Figure 1.1: Relative domains of typical frequency shifts for Rayleigh, Rayleigh-wing, Brillouin, and Raman scattering.

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.[1] 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[2], just one year after the first demonstration of the laser by American physicist Theodor Maiman.[3] 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 stimulated brillouin scattering process, and Raman scattering.

1.1 Spontaneous Light Scattering

Light scattering involves the redirection of light as a result of interactions with the constituent particles or molecules within a material medium. In every case, light scattering occurs because of variations in the material's optical properties. To understand why, envision a material with completely uniform particles—spatially and temporally consistent, or in other words, perfectly homogeneous. Figure 1.2 shows an incident optical plane wave encountering a segment of such a material, denoted δz, containing a volume element δV_1. For any given incident wavelength λ and any non-zero scattering angle θ at volume δV_1, there exists a corresponding volume element δV_2, located a distance $\frac{\lambda}{2\sin\theta}$ apart, which scatters light at the same angle θ. The scattered waves from δV_1 and $\delta V2$ 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.[1] 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 treating the higher-intensity stimulated Brillouin scattering and ultimately I will present the coupled-wave equations of Coherent Anti-Stokes Brillouin Spectrometer (CABS), the instrument which underpins many of my results. Let us build a theoretical description considering thermodynamic fluctuations as the origin of the light scattering process.

1.2 Spontaneous Brillouin Scattering

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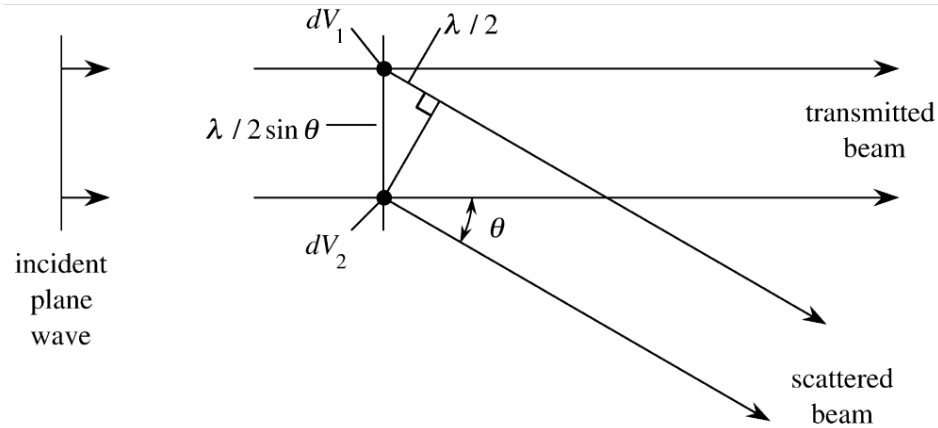

Figure 1.2

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1.3 Stimulated Brillouin Scattering

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1.4 Phase-matching

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1.5 Brillouin Gain of Materials

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1.6 Raman Scattering

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1.7 Raman-like Brillouin Modes

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Chapter 2

Foundational Experimental Techniques and Instrumentation

This is an inline citation,[1]. This is a parenthetical citation[1]. This is a figure reference (Figure 1.1). This is a section reference §1.2. This is a chapter reference with chapter spelled out: chapter 3. This is an acronym definition **AGU!** (**AGU!**). This is the second time I use the acronym in this section **AGU!**. This is if I want to spell out the full acronym again **AGU!** (**AGU!**). Define new acronyms in the acronyms.tex file.

2.1 Experimental Techniques

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2.1.1 ways we can direct light in a photonic system

2.1.2 photonic devices and diagrams

2.1.3 ways we can select and isolate signals

2.1.4 heterodyne detection and the role of the LO

2.1.5 loss in a photonic system

2.1.6 free space optics and beam alignment

2.1.7 special fiber types and properties

2.2 Optical Instrumentation

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2.3 Electronic Instrumentation

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2.4 Noise and Background Handling

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2.5 Custom Software

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2.5.1 Description of Python Script for CABS Data Collection

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2.5.2 Description of Plotting Data in Go Program

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Chapter 3

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

3.1 Abstract

3.2 Optomechanical Cooling and Heating

3.3 Cooling Platform: CS_2-Liquid Core Optical Fiber

3.3.1 Optomechanical Properties

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3.3.3 Fabrication Iterative Refinement

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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 Syncronous Achievement by Max Plank Group

3.7.3.1 Platform: Tapered chalcogenide Photonic Crystal Fiber

Chapter 4

Manuscript II: A coherently stimulated phonon spectrometer

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

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4.1 Abstract

4.2 Introduction

State of brillouin microscopy Applications and usefulness Challenges: selection of backscattered signal conflated with Stokes field phase-matching requires probe wavelength to be exactly that of Stokes Wouldn't it be nice if we could break free of strict phase-matching requirements, therefore perfectly isolating the signal In this work

4.3 Instrument Design

4.3.1 Design of instrument

description of design figure: instrument apparatus design

4.3.2 Sensitivity Measurements

4.4 Theory

4.4.1 Coupled Wave Equations

full CABS theory arriving at scattered power

[1] Department of Applied Physics and Materials Science, Northern Arizona University, Flagstaff, AZ 86011, USA
[2] Center for Materials Interfaces in Research and Applications, Flagstaff, AZ 86011, USA
[3] Sandia National Laboratory, 1515 Eubank Blvd SE, Albuquerque, NM 87123, USA
[4] Department of Applied Physics, Yale University, New Haven, CT 06520, USA

4.4.2 Phase-matching bandwidth

phase-matching bandwidth theory

4.5 Results

4.5.1 Fiber-Coupled: UHNA3

4.5.2 Free-Space: CS_2

figure: demonstration measurements 1mm uhna3 fiber 1mm CS2 bulk

comparison to stimulated brillouin and spontaneous brillouin?

4.5.3 Phase-Matching in Small L Regime

figure: phase-matching peak vs pump-probe separation 1cm uhna3, CS2 peak vs pump-probe separation 1mm uhna3, CS2

4.6 Discussion

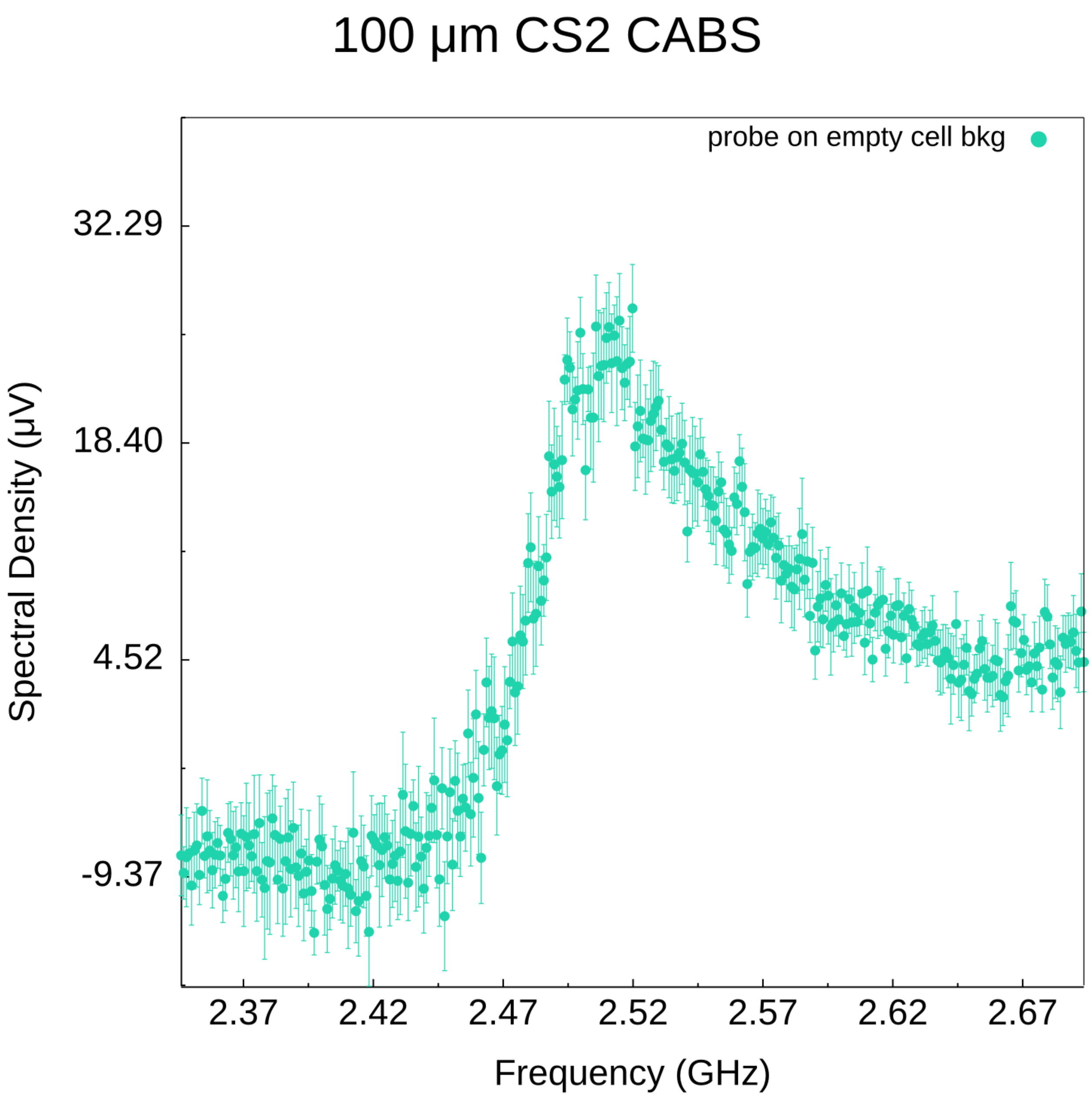

Figure 4.1: CABS measurement of 100um of CS2.

Chapter 5

Manuscript III: Brillouin-induced Raman modes

5.1 Abstract

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5.2 Introduction

Nam dui ligula, fringilla a, euismod sodales, sollicitudin vel, wisi. Morbi auctor lorem non justo. Nam lacus libero, pretium at, lobortis vitae, ultricies et, tellus. Donec aliquet, tortor sed accumsan bibendum, erat ligula aliquet magna, vitae ornare odio metus a mi. Morbi ac orci et nisl hendrerit mollis. Suspendisse ut massa. Cras nec ante. Pellentesque a nulla. Cum sociis natoque penatibus et magnis dis parturient montes, nascetur ridiculus mus. Aliquam tincidunt urna. Nulla ullamcorper vestibulum turpis. Pellentesque cursus luctus mauris.

5.3 Methods

Nam dui ligula, fringilla a, euismod sodales, sollicitudin vel, wisi. Morbi auctor lorem non justo. Nam lacus libero, pretium at, lobortis vitae, ultricies et, tellus. Donec aliquet, tortor sed accumsan bibendum, erat ligula aliquet magna, vitae ornare odio metus a mi. Morbi ac orci et nisl hendrerit mollis. Suspendisse ut massa. Cras nec ante. Pellentesque a nulla. Cum sociis natoque penatibus et magnis dis parturient montes, nascetur ridiculus mus. Aliquam tincidunt urna. Nulla ullamcorper vestibulum turpis. Pellentesque cursus luctus mauris.

5.4 Results

Nam dui ligula, fringilla a, euismod sodales, sollicitudin vel, wisi. Morbi auctor lorem non justo. Nam lacus libero, pretium at, lobortis vitae, ultricies et, tellus. Donec aliquet, tortor sed accumsan bibendum, erat ligula aliquet magna, vitae ornare odio metus a mi. Morbi ac orci et nisl hendrerit mollis. Suspendisse ut massa. Cras nec ante. Pellentesque a nulla. Cum sociis natoque penatibus et magnis dis parturient montes, nascetur ridiculus mus. Aliquam tincidunt urna. Nulla ullamcorper vestibulum turpis. Pellentesque cursus luctus mauris.

5.5 Discussion

Nam dui ligula, fringilla a, euismod sodales, sollicitudin vel, wisi. Morbi auctor lorem non justo. Nam lacus libero, pretium at, lobortis vitae, ultricies et, tellus. Donec aliquet, tortor sed accumsan bibendum, erat ligula aliquet magna, vitae ornare odio metus a mi. Morbi ac orci et nisl hendrerit mollis. Suspendisse ut massa. Cras nec ante. Pellentesque a nulla. Cum sociis natoque penatibus et magnis dis parturient montes, nascetur ridiculus mus. Aliquam tincidunt urna. Nulla ullamcorper vestibulum turpis. Pellentesque cursus luctus mauris.

Nam dui ligula, fringilla a, euismod sodales, sollicitudin vel, wisi. Morbi auctor lorem non justo. Nam lacus libero, pretium at, lobortis vitae, ultricies et, tellus. Donec aliquet, tortor sed accumsan bibendum, erat ligula aliquet magna, vitae ornare odio metus a mi. Morbi ac orci et nisl hendrerit mollis. Suspendisse ut massa. Cras nec ante. Pellentesque a nulla. Cum sociis natoque penatibus et magnis dis parturient montes, nascetur ridiculus mus. Aliquam tincidunt urna. Nulla ullamcorper vestibulum turpis. Pellentesque cursus luctus mauris.

Table 5.1: Table caption.

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

Chapter 6

Discussion & Future Work

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Appendix A

Acronyms

SBS Stimulated Brillouin Scattering

CABS Coherent Anti-Stokes Brillouin Spectrometer

Appendix B

Code

B.1 Python Code for CABS Data Collection

B.2 Plotting Data In Go Program

Appendix C

Supplementary Information for Chapter 3: Manuscript I

C.1 Data

Appendix D

Supplementary Information for Chapter 4: Manuscript II

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

figure: P, S, Pr equal contributors

D.2 Data

Appendix E

Supplementary Information for Chapter 5: Manuscript III

E.1 Data

References

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[2] eg PA Franken, Alan E Hill, CW el Peters, and Gabriel Weinreich. Generation of optical harmonics. *Physical review letters*, 7(4):118, 1961.

[3] Theodore H Maiman et al. Stimulated optical radiation in ruby. 1960.