High Pressure Phase Diagram of Silicon

by

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Biographical Sketch

The author was born in Danyang, Jiangsu Province, China. He attended University of Science and Technology of China, and graduated with a Bachelor of Science degree in Applied Physics. He began doctoral studies in Department of Mechanical Engineering at the University of Rochester in 2014. He was awarded the Horton Fellowship and received the Master of Science degree from the University of Rochester. He pursued his research in High Energy Density Physics under the direction of Ryan Rygg and Gilbert Collins.

The following publications were a result of work conducted during doctoral study:


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I have had the opportunity to collaborate with scientists from outside the LLE. In particular, I am grateful for the guidance from Dr. Amy Jenei, Dr. Ray Smith, Dr. Jon
Eggert, Dr. Federica Coppari from Lawrence Livermore National Laboratory (LLNL). I would like to thank Jon Eggert and Marius Millot at Lawrence Livermore National Laboratory for the permission to use AnalyzeVISAR and AnalyzePXRDIP analysis software packages.
The experiment described in this thesis explores the phase diagram of silicon near its isentrope from 40 to 400 GPa, by ramp compressing silicon by a laser drive. Thermodynamic states of silicon at these states are measured by velocimetry, and the crystal structure is determined by nanosecond in-situ x-ray diffraction. The experiment shows a significant increase of the stability range of the Si hcp phase compared to theoretical predictions. The hcp phase is observed at the pressure and temperature range where dhcp phase was predicted, and no evidence of the dhcp phase is observed. Furthermore, the hcp-fcc phase transition pressure is at least 93 GPa, much higher than the 55 GPa predicted by computation. This observation is consistent with previous shock compression experiments. The fcc phase is confirmed to remain stable to at least 400 GPa.

Currently, no temperature data exist from nanosecond pyrometric measurements on ramp compression experiments. Such measurements are difficult due to the low number of photons emitted from low temperature (lower than 4000 K) targets. In this work, we present the foundational framework for analyzing low signal-to-noise ratio data. This method yields identical results as traditional techniques at high temperatures, but is more robust at low temperatures. This sets the stage for analyzing future low temperature pyrometry data.
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Chapter 1

Introduction

High-energy-density (HED) physics [6–8] is the field of physics studying matter under extreme states of pressure. Energy density and pressure are usually synonymous, except in the context of the general theory of relativity. The standard definition of HED is energy densities above 100 GPa (or equivalently 1 million of Atmosphere, or 1 Mbar) [9] but some authors like to refer to HED by \( P > 10 \) GPa [8].

At HED conditions, pressure can approach that of the electrons, and the interatomic distance can be shorter than the de Broglie wavelength of ions. For example, the typical energy of a chemical bond is of the order of 1 eV, and the length of the bond is \( \sim 1 \) Å, so the energy density of a typical chemical bond is \( \sim 1 \text{eV}/(1 \text{Å})^3 \approx 1.6 \times 10^{11} \text{J/m}^3 = 160 \) GPa. Even at pressures lower than this number, the chemical properties can be drastically changed due to the pressure. For another example, at 1000 K, the de Broglie wavelength of a deuterium atom is \( \lambda = \frac{h}{\sqrt{3Am_uk_BT}} \approx 0.56 \) Å. The density of a deuterium system whose interatomic distance equals \( \lambda \) is \( \sim 4.4 \text{g/cm}^3 \). When the de Broglie wavelengths of ions are comparable to the interatomic distances, even the motion of the ions must be described quantum-mechanically, and macroscopic quantum phenomenon, such as superfluidity and superconductivity, can emerge.

Examples of places where these conditions occur in nature include normal stars, exploding stars, active galaxies, and planetary interiors. Human-generated HED matter has been created since the days of World War II, and later in the laboratory,
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revealing altogether new states of matter such as high-pressure elemental electrides [4], metallic fluid hydrogen [10], and super ionic water [11]. HED physics is, by its nature, an interdisciplinary field that encompasses condensed matter physics, plasma physics, astrophysics, and planetary science, among others.

This thesis studies the high-pressure phase diagram of silicon. Silicon is commonly used in semiconductor industry, and is an important component of Earth-like planets. Earth’s core is composed of iron alloyed with nickel and some amount of lighter elements such as H, C, S, O, and Si [12]. The understanding of the behavior of Earth-like planets is deepened by studying the phase diagram of silicon alone. The phase diagrams of diamond [13] and tin [14], both in the same group as Si, have been studied previously, showing unexpected behavior compared to theoretical predictions. The phase diagram of silicon has recently been surveyed with evolutionary phase search algorithms [15, 16], providing a state of the art prediction for its high pressure behavior [17–19]. This thesis presents measurements of the Si phase diagram at pressures between 40 and 400 GPa and densities between 4 and 7 g/cm³, providing rigorous benchmarks for theoretical predictions.

The HED conditions can be created by static means, such as diamond anvil cell (DAC) [20], or dynamic means, such as gas gun [21], pulsed power devices like Z machine [22], and high-power lasers like OMEGA [23], OMEGA EP [24], and NIF [25]. At OMEGA EP facility, matter is compressed by direct laser illumination. Matter is rapidly heated by high-power laser such that it blows off or ablates, creating a reaction force which drives a pressure wave into the sample. Such ablation produced pressures are large enough that even solids are significantly compressed. While the motion of such pressure waves is described by hydrodynamics in fluids, we use hydrodynamic formalism to largely describe such pressure waves in solids also, however the effects of material strength and anisotropy need to be included (Section 2.2). The experimental study of the phase diagram requires measurement of pressure, density, and temperature. Pressure is determined by hydrodynamic means, and the
principles behind this measurement are discussed in Sections 2.3 and 3.2. Whether the sample is still solid and what crystal structure the sample assumes are answered by means of x-ray diffraction, as discussed in Sections 2.5 and 3.1. In Chapter 4, a measurement of phase diagram of silicon from 40 to 400 GPa near the isentrope is presented. Finally, the temperature is measured using the black-body radiation of the sample, whose principles are discussed in Section 3.3. Currently the temperature measurement does not yield satisfactory results due to the low-temperature nature of the thermodynamic states of interest. In Chapter 5, we present an effort to extend the temperature measurement to low-temperature regimes.
Chapter 2

Theory

2.1 Equation of State (EOS)

A subsection of HEDP studies the equations of state of matters at high-energy-density conditions. Equilibrium matter obeys the universal laws of thermodynamics. In addition, each material has its own characteristic thermodynamic features distinct from other materials, and these are described by its equations of state.

Formally, equations of state are relationships expressing intensive parameters (such as temperature $T$, pressure $P$, density $\rho$) in terms of independent extensive parameters (such as entropy $S$, volume $V$) \[26\]. The Los Alamos SESAME Database EOS Library is the most used EOS database among the HEDP community. It includes tables of pressure $P$ and internal energy $E$ (also of Helmholtz free energy, $A$ sometimes), each as a function of density $\rho$ and temperature $T$. Various other EOS database has been constructed in a similar format as SESAME, such as LEOS, FPEOS, etc.

In SESAME, each material is represented by one or more files. Multiple files may exist for one material, each representing a different modeling of the material. In each file, there are multiple tables containing various aspect of information of the material. Table 101 is a short description of the material; table 102 is a long description of the table and the modeling method used to generate the table; table 103 is similar to table 102. Table 201 contains five basic parameters of the material, of
which the first three are crucial: mean atomic number $Z_{\text{BAR}}$, mean atomic mass $A_{\text{BAR}}$, normal (solid) density $\rho_{00}$. Table 301 is the total EOS table, which is what is usually used in practice. The first two entries of the table are the number of densities $N_R$ and the number of temperatures $N_T$. The next $N_R$ entries are the density array $R$, and the $N_T$ entries that follow are the temperature array $T$. The next $N_R \cdot N_T$ entries form the pressure array $P$, and the indexing scheme is $(P(I, J), I=1, N_R), J=1, N_T)$. The next $N_R \cdot N_T$ entries form the internal energy array $E$ with the same indexing scheme. Some 301 tables end here, but some will contain another array, namely the Helmholtz free energy array $A$ with another $N_R \cdot N_T$ entries. A SESAME file might contain other EOS tables. Table 303, 304, 305 are EOS tables of ion plus cold curve, electron alone, and ion alone, respectively. They are the component tables of table 301, occasionally useful when one wants to examine the contribution to pressure or internal energy from each constituent.

In HED physics, frequently one needs to compute the isentrope or Hugoniot of a material using an EOS table. An isentrope is a thermodynamic path along which the entropy is kept constant. The isentrope whose entropy is the entropy at ambient conditions is called the principal isentrope. The isentrope can be found by interpolating the entropy array to find pairs of densities and temperatures $(\rho_i, T_i)$ of constant entropy. This is easy if the SESAME file contains the Helmholtz free energy table $A$, for entropy can be computed by $S = (A - E)/T$. If the SESAME file does not contain the Helmholtz free energy table, one can integrate the differential relation $dE = -PdV = (P/\rho^2)d\rho$ to find the pairs $(\rho_i, T_i)$, which is slightly harder because it requires interpolating two tables $E$ and $P$ simultaneously.

A Hugoniot is the locus of all end states achieved via a single shock wave from the same state. The phenomena of shock waves are briefly mentioned in Section 2.3 and discussed extensively in [27]. In the case of a one-dimensional steady shock (Figure 2.1), the state $\rho_0, v_0, P_0, E_0$ variables of the unshocked material are assumed to be known, and the strength of the shock wave can be described by pressures of
Figure 2.1: Diagram of an isolated, steady shock, in the laboratory reference frame and a frame that moves with the shock (shock frame), respectively. In the laboratory frame, the unshocked material and shocked material moves at \( v_0 \) and \( v_1 \), and the shock front moves with a speed \( D \) (\( D > v_0, v_1 \)). In the shock frame, the unshocked material moves towards the shock front with a speed \( D - v_0 \) while the shocked material moves away from the front with a speed \( D - v_1 \). The thermodynamic quantities \( P, \rho, E \) on both sides are invariant under the Galilean transformation.

the shocked material \( R_1 \), or the particle velocity discontinuity \( v_1 - v_0 \). The Hugoniot is described by three conservation laws,

\[
\begin{align*}
\rho_1 u_1 &= \rho_0 u_0, \\
P_1 + \rho_1 u_1^2 &= P_0 + \rho_0 u_0^2, \\
E_1 + \frac{P_1}{\rho_1} + \frac{u_1^2}{2} &= E_0 + \frac{P_0}{\rho_0} + \frac{u_0^2}{2},
\end{align*}
\] (2.1)

where \( u_0 = D - v_0 \) is particle velocity relative to shock front. For each specified \( u_1 \), the particle velocity after the shock, one can solve these equations to obtain relations \( \rho_1(u_1), P_1(u_1), E_1(u_1) \). By eliminating \( u_1 \), one obtains

\[
\begin{align*}
P_1 &= P_0 + \rho_0 u_0^2 \left( 1 - \frac{\rho_0}{\rho_1} \right), \\
E_1 &= E_0 + P_0 \left( \frac{1}{\rho_0} - \frac{1}{\rho_1} \right) + \frac{u_0^2}{2} \left( 1 - \frac{\rho_0}{\rho_1} \right)^2.
\end{align*}
\] (2.2)

This is the format suitable for interpolation.

During this thesis work, I have written a Python package to read SESAME tables
and compute isentropes and Hugoniots given initial conditions. The code is essentially a simplified Python version of EOSPAC package [28]. The code in particular uses a birational interpolation scheme [29] to avoid overshooting and wrong sign of slopes of the interpolated EOS tables. This is especially important when one wishes to compute the derivatives from the table. Examples of such derivatives include the sound speed, the specific heat, the Grüneisen coefficient, etc. The code have a few low-level interpolation functions and two high level classes Isentrope and Hugoniot. The two classes can be initiated by specifying initial density and temperature. If initial pressure and either initial density or temperature is known, then the other variable can be found by using one of the low-level interpolators. Both classes have methods that transform one of \((\rho, T, P)\) quantities to the other two along an isentrope or Hugoniot.

### 2.2 Equations of Hydrodynamics

Materials that are dynamically compressed to extreme conditions may not be in global thermodynamic equilibrium. However, any volume element in the macroscopic body that is small enough compared to the size of the macroscopic body, but still large enough to contain a great number of atoms, can be regarded as in local equilibrium at a given instant. Therefore the thermodynamic properties of macroscopic bodies can be described in terms of the thermodynamic properties of these small-volume fluid elements. In other words, macroscopic bodies are treated as continuous media. In the high-energy-density regime, even solids can undergo significant compression, thus the equations for compressible fluids can often be used as the governing equations.

The two approaches for the treatment of compressible flow, Lagrange or Eulerian, differ in the reference frame considered, i.e., on moving with a fluid element or a fixed control volume. Consider a fluid element, located at \(X\) at some reference time \(t = 0\), moves to a new position \(x\) at a later time \(t\). Each fluid element can be
distinguished from others by its unique initial location \( \mathbf{X} \), so \( \mathbf{x} \) is a function of \( t \) and \( \mathbf{X} \), and the mechanical motion of the macroscopic body may be represented by \( \mathbf{x} = \mathbf{x}(\mathbf{X}, t) \). Knowing the time evolution \( F_L(\mathbf{X}, t) \) of the thermodynamic quantity \( F \) of each element \( \mathbf{X} \) is equivalent to knowing the time evolution of \( F \) of the entire macroscopic body. This is the Lagrangian point of view, and \( \mathbf{X} \) is usually called the Lagrangian coordinate. If the motion is continuous and single-valued, it is possible to solve for \( \mathbf{X} = \mathbf{X}(\mathbf{x}, t) \). The thermodynamic quantity \( F \) of the entire macroscopic body can then be expressed as a function of \( \mathbf{x} \) and \( t \) by \( F_E(\mathbf{x}, t) = F_L(\mathbf{X}(\mathbf{x}, t), t) \). \( F_E(\mathbf{x}, t) \) is a field, i.e., a function of space and time, just like the familiar electromagnetic fields. This is the Eulerian point of view, and \( \mathbf{x} \) is sometimes called the Eulerian coordinate. The subscripts \( E \) and \( L \) are to emphasize which point of view is used; the functions \( F_E \) and \( F_L \) are numerically identical (this is similar to the difference between the energy function \( E(q, \dot{q}) \) expressed in terms of generalized coordinate \( q \) and generalized velocity \( \dot{q} \) and the Hamiltonian \( H(q, p) \) expressed in terms of canonical variables in mechanics.)

It is customary to write the time derivative of \( F \) in Eulerian and Lagrangian frames in the following way:

\[
\frac{\partial F}{\partial t} := \frac{\partial F_E}{\partial t}, \quad \frac{DF}{Dt} := \frac{\partial F_L}{\partial t} = \frac{\partial F_E}{\partial t} + \frac{d}{dt} \frac{\partial F_E}{\partial \mathbf{x}_i} = \frac{\partial F}{\partial t} + \mathbf{u}_i \frac{\partial F}{\partial \mathbf{X}_j} \frac{\partial \mathbf{X}_j}{\partial \mathbf{x}_i}, \tag{2.3}
\]

where \( \mathbf{u} \) is the particle velocity, and the spatial derivatives are related by

\[
\frac{\partial F}{\partial \mathbf{x}_i} = \frac{\partial F_E}{\partial \mathbf{x}_i} = \frac{\partial F_L}{\partial \mathbf{X}_j} \frac{\partial \mathbf{X}_j}{\partial \mathbf{x}_i}. \tag{2.4}
\]

The complete set of equations of motion, ignoring body forces and heat conduc-
tion, is \[30\]

\[
\frac{D\rho}{Dt} + \rho \frac{\partial u_i}{\partial x_i} = 0 \quad \text{(conservation of mass)}
\]

\[
\frac{Du_i}{Dt} - \frac{1}{\rho} \frac{\partial \sigma_{ji}}{\partial x_j} = 0 \quad \text{(Newton’s second law)}
\]

\[
\frac{D\varepsilon}{Dt} + \frac{\sigma_{ji}}{\rho} \frac{\partial u_j}{\partial x_i} = \dot{S} \quad \text{(first law of thermodynamics)}
\]

(2.5)

where \(\sigma_{ji}\) is the stress tensor, related to pressure by

\[
\sigma_{ij} = -p\delta_{ij} + \tau_{ij},
\]

(2.6)

\(\tau_{ij}\) is the deviatoric stress tensor whose trace is zero, \(\varepsilon\) is the internal energy density, and \(\dot{S}\) is the rate of internal heat generation per unit mass.

In the kind of experiments described in this thesis, the motion of a compressed sample is almost planar, where all functions depend only on one Cartesian variable \(x\). In this case, the stress tensor \(\sigma_{ij}\) is diagonal by symmetry. If the fluid is initially at rest and the initial density is uniform, \(\rho(x, 0) = \rho_0\), then \(X = X(x, t)\) becomes the differential relation \(\rho_0 \, dx = \rho \, dx\). The three equations take the form

\[
\frac{D\rho}{Dt} + \frac{\rho^2}{\rho_0} \frac{\partial u}{\partial X} = 0,
\]

\[
\frac{Du}{Dt} - \frac{1}{\rho_0} \frac{\partial \sigma_x}{\partial X} = 0,
\]

(2.7)

\[
\frac{D\varepsilon}{Dt} + \frac{\sigma_x}{\rho_0} \frac{\partial u}{\partial X} = \dot{S}.
\]

### 2.3 Method of Characteristics

If the flow is devoid of shock waves and other entropy generation mechanisms such as plastic flow, the flow is adiabatic, \(\dot{S} = 0\). The density \(\rho\) is related to stress \(\sigma_x\) and entropy \(S\) by the equation of state \(\rho = \rho(\sigma_x, S)\), and since \(dS/dt = 0\), we have

\[
\frac{D\rho}{Dt} = \left(\frac{\partial \rho}{\partial \sigma_x}\right) \frac{D\sigma_x}{Dt} = \frac{1}{c_E^2} \frac{D\sigma_x}{Dt} = \frac{\rho^2}{\rho_0^2} \frac{1}{c_L^2} \frac{D\sigma_x}{Dt}.
\]

(2.8)
where $c_E$ and $c_L$ are the Eulerian sound speed and Lagrangian sound speed,

$$c_E = \sqrt{\left( \frac{\partial \sigma_x}{\partial \rho} \right)_S}, \quad c_L = \frac{\rho}{\rho_0} c_E,$$

respectively. Combining the mass and momentum equation, we obtain

$$\left[ \frac{D u}{D t} \pm c_L \frac{\partial u}{\partial X} \right] \pm \frac{1}{\rho_0 c_L} \left[ \frac{D \sigma_x}{D t} \pm c_L \frac{\partial \sigma_x}{\partial X} \right] = 0 \quad (2.10)$$

The quantities in the brackets can be viewed as the directional derivative of $u$ and $\sigma_x$ along certain curves in the $t - X$ plane, called the characteristics. Two families of characteristics exist for this equation,

$$C_\pm : \quad \frac{d X}{d t} = \pm c_L. \quad (2.11)$$

The infinitesimal increment of $u$ along $C_\pm$ is

$$d u = \frac{D u}{D t} dt + \frac{\partial u}{\partial X} dX = \left( \frac{D u}{D t} + \frac{dX}{dt} \frac{\partial u}{\partial X} \right) dt = \left[ \frac{D u}{D t} \pm c_L \frac{\partial u}{\partial X} \right] dt. \quad (2.12)$$

Similar relations hold for $\sigma_x$. Equation (2.10) is reduced to

$$d u \pm \frac{1}{\rho_0 c_L} d \sigma_x = 0 \quad \text{along} \quad C_\pm. \quad (2.13)$$

If the flow, in addition to being adiabatic, is isentropic, i.e., the entropy is constant both spatially and temporally, then any one of the thermodynamic variables $\rho$, $\sigma_x$, or $c_L$ are functions of each other. Figure 2.2 shows the principal isentrope of diamond to 800GPa, along which the entropy $S$ is kept constant. In panels (a) and (b), the stress $\sigma_x$ and Lagrangian sound speed $c_L$ are shown as functions of the density $\rho$. By eliminating $\rho$, the stress $\sigma_x$ can be expressed as a function of $c_L$. 
Equation (2.13) shows that the quantities

\[ J_\pm = u \pm \phi, \]  

(2.14)
called Riemann invariants, are constants along each characteristic, where

\[ \phi(\sigma_x) = \int \frac{d\sigma_x}{\rho_0 c_L} \]  

(2.15)
is a function of any one of the variables \( \rho, \sigma_x, \) or \( c_L. \)

By solving Equation (2.14), \( u \) and \( c_L \) can be expressed by \( J_+ \) and \( J_- \), so the equations for characteristics Equation (2.11) can be expressed as

\[ C_+ : \frac{dX}{dt} = F_+(J_+, J_-); \quad C_- : \frac{dX}{dt} = F_-(J_+, J_-). \]  

(2.16)

If for some reason \( J_- \) is constant spatially and temporarily, then the slope of each \( C_+ \) depends on the \( J_+ \) of that particular \( C_+ \), which is a constant along \( C_+ \). Therefore each \( C_+ \) is a straight line whose slope is determined by the Riemann invariant \( J_+ \) carried along it. It is evident that along each \( C_+ \), the functions \( u \) and \( c_L \) are constants, but the constants for different \( C_+ \) may differ. Therefore, the solution to the hydrodynamic equations takes the form

\[ u = f(X - c_L(u)t), \quad c_L = g(X - c_L(u)t). \]  

(2.17)
The explicit form of the functions \( f \) and \( g \) is determined by the boundary and initial conditions. This solution has the form of a traveling wave, where the wave speed \( c_L(u) \) depends on the local particle velocity. This type of solution is called a simple wave. The initial profiles of \( u(X, 0) \) and \( c_L(X, 0) \) can become distorted with time, giving rise to the phenomenon of shock waves.
2.3.1 Planar Motion Induced by an Applied Stress

Consider a sample occupying the half space $X \geq 0$. Suppose at the initial time $t = 0$, the sample is at rest and relaxed, so the initial conditions are $u(X, 0) = 0$ and $\sigma_x(X, 0) = 0$ for every $X \geq 0$. The $C_-$ characteristics emanating from the positive $X$-axis, identified by $X$ where they originate at $t = 0$, carry along them the Riemann invariant

$$J_-(X) = u(X, 0) - \phi(\sigma_x(X, 0)) = 0.$$  \hspace{1cm} (2.18)

Therefore $J_- = 0$ for every $X \geq 0$ and $t \geq 0$, and the solution is a simple wave. For all $X \geq 0$ and $t \geq 0$, $u(X, t)$ and $\phi(X, t)$ are constrained by

$$J_- = u(X, t) - \phi(X, t) = 0,$$  \hspace{1cm} (2.19)

so the purely kinematic quantity $u$ joins the rank of thermodynamic quantities $\rho$, $\sigma_x$, $c_L$, and $\phi$, in the sense that each of them can be expressed as a function of another. In Figure 2.2(c), particle velocity $u$ and the function $\phi$ is shown as functions of $\rho$. They can also be expressed as functions of $\sigma_x$ and $c_L$.

At $t = 0$, a time-varying stress $\sigma_{x0}(t)$ is applied on the sample at $X = 0$, and the sample starts to move. The characteristics $C_+$ emanating from the positive $t$-axis carry along them can be identified by $t$ where they originate at $X = 0$. Each carries its Riemann invariant:

$$J_+(t) = u(0, t) + \phi(\sigma_{x0}(t)).$$  \hspace{1cm} (2.20)

The equation for the characteristic $C_+(t')$ is

$$X = c_L[\sigma_{x0}(t')](t - t'),$$  \hspace{1cm} (2.21)
Figure 2.2: Experimentally measured principal isentrope of diamond up to 800 GPa [31]. Along an isentrope, the entropy $S$ is kept constant, so thermodynamic variables are functions of each other. In particular, (a) shows stress $\sigma_x$ as a function of density $\rho$, and (b) shows the Lagrangian sound speed $c_L$ as a function of $\rho$. (c) For simple waves, the particle velocity $u$ and the integral $\phi$ are also functions of $\rho$, and $u = \phi$ numerically if the integration constant is chosen as zero in Equation (2.15).
a straight line. Along \( C_+(t') \),

\[
u(X, t) = \phi(X, t) = \frac{1}{2} J_+(t') = \frac{1}{2} [u(0, t') + \phi(\sigma_{x0}(t'))] = \phi[\sigma_{x0}(t')]. \tag{2.22}
\]

The complete solution \( u(X, t) \) (and thus other quantities \( \sigma(X, t) \), etc., since they are functions of each other) is obtained by eliminating \( t' \) from Equations (2.21) and (2.22).

**Example Problem Solved with Characteristics**

Consider the compression of a diamond sample with an equation of state shown in Figure 2.2. In this problem the applied stress \( \sigma_{x0} \) increases quadratically with time, and reaches 400 GPa at 10 ns, then \( \sigma_{x0}(t) = 4(t\text{[ns]})^2 \text{GPa} \) (see Figure 2.3a). We discretize the \( t \) axis and denote the \( C_+ \) characteristics emanating from these points as \( C_+(0), C_+(1) \), etc, and the \( C_- \) characteristics arriving at these points as \( C_-(0), C_-(1) \), etc. The intersection of \( C_+(i) \) and \( C_-(j) \) is denoted by \( (i, j) \). The boundary condition \( \sigma_x(0, t) = \sigma_{x0}(t) \) is implemented by assigning the Riemann invariant for \( C_+(i) \) as \( J_+(i) = 2\phi[\sigma_{x0}(t_i)] \). The particle velocity and \( \phi \) at the intersection \( (i, j) \) are \( u^{i,j} = \phi^{i,j} = J_+(i)/2 = \phi[\sigma_{x0}(t_i)] \). The stress and sound speed at \( (i, j) \) can be obtained by the relations \( \sigma_x = \sigma_x(\phi), c_L = c_L(\phi) \). The flow variables at all intersections have been obtained, but the locations of these intersections are still unknown. They can be obtained by solving the equations

\[
x^{i,j} = x^{i,j-1} + c_p(t^{i,j} - t^{i,j-1}) \tag{2.23}
\]

\[
x^{i,j} = x^{i+1,j} - c_m(t^{i+1,j} - t^{i,j}).
\]

where

\[
c_p = \frac{1}{2}(c_{L_{i,j}} + c_{L_{i,j-1}}), \quad c_m = \frac{1}{2}(c_{L_{i,j}} + c_{L_{i+1,j}}) \tag{2.24}
\]
by the following order

\[(0, 0) \rightarrow (1, 1) \rightarrow (2, 2) \cdots \rightarrow (0, 1) \rightarrow (1, 2) \rightarrow (2, 3) \cdots \rightarrow (0, 2) \rightarrow (1, 3) \rightarrow (2, 4) \cdots \rightarrow \vdots \quad (2.25)\]

The characteristics of said motion are shown in Figure 2.3b. The $C_+$ and $C_-$ characteristics are shown in red and blue, respectively, and the indexing scheme of the intersections should be self-evident. As stated, the $C_+$ characteristics are all straight lines with constant slopes. The slopes of the $C_-$ characteristics are constant until they intersect the $C_+$ characteristics.

The region of solution is the “triangular” region enclosed by the positive $t$ axis, the first $C_+$ characteristic ($C_+(0)$ in Figure 2.3b) and the last $C_-$ characteristic ($C_-(6)$ in Figure 2.3b), where the solution is completely determined by the applied boundary condition. Therefore the spatial extent of the region of solution is determined by the length of temporal input. In Figure 2.3(c), the temporal input is from 0 ns to 15 ns, and the spatial extent of the solution is from 0 $\mu$m to 140 $\mu$m. If knowing what happens beyond 140 $\mu$m is desired, one can often pad the temporal input as appropriate. In this example, it is known that the applied stress is zero both before 0 ns and after 10 ns, so the applied stress can be extended temporally by appending zeros at both ends. Comparing Figure 2.3(c) and (d), one can clearly see the improvement of the spatial extent of the solution if one pads the input applied stress, from 140 $\mu$m to 280 $\mu$m.

The improvement is however not without limit. The $C_+$ characteristics started from 5 ns to 10 ns converge and overlap with each other at around 180 $\mu$m. This is because the sound speed is an increasing function of stress at $\sigma_x > 100$ GPa (Figure 2.2) and the applied stress is an increasing function of time. The slope of
Figure 2.3: An example of planar motion induced by an applied stress. The half space $X \geq 0$ is filled with diamond initially at rest. (a) The stress applied at $X = 0$ is $\sigma_{\infty}(t) = 4(t[\text{ns}])^2 \text{GPa}$ for $0 \leq t \leq 10 \text{ ns}$ and 0 otherwise. (b) Six $C_+$ characteristics (red) and six $C_-$ characteristics (blue), and the indexing scheme. (c) The solution of the problem if the input boundary condition lasts 15 ns. Characteristics with 1 ns distances as well as stress solution at the intersections of $C_-$’s and $C_+$’s are shown. (d) Same as (c), except that the input boundary condition lasts 30 ns. (e) If the sample has finite thickness, only solutions over a finite region in the $t$-$X$ planes can be uniquely determined using the current algorithm (the blue), while the previously valid region of solution (red) has been invalidated by the existence of the sample back-surface.
the $C_+$ characteristics is $c_L$, increasing with time, so the subsequent $C_+$ have larger and larger slopes, until they take over earlier ones. This will lead to multivalued solutions to the hydrodynamic equations, which is not physical. In reality, what happens is that the solution becomes discontinuous and this treatment breaks down. The discontinuous solution is a shock wave, which is discussed extensively in [27]. The method of characteristics does not handle the situation where a shock wave emerges.

This scheme assumes that the sample occupies the entire half space $X > 0$, while in reality samples have finite thicknesses. The region of determination in the case of a sample of finite thickness is enclosed by the positive $t$ axis, the first $C_+$ characteristic, and the first “reflected” $C_-$ characteristic “reflected” from the sample surface (see the blue region in Figure 2.3e). Although the boundary condition may be applied for a long period, any information after the point where the first reflected $C_-$ characteristic intersects the $t$ axis is not suitable as the boundary condition for this problem. Therefore, in practical situations one must pay attention to the “topology” of the characteristics. This problem is solvable, but the scheme for the method of characteristics needs to be modified and is not covered here.

2.3.2 Infer a Planar Motion from Measured Interface Velocity

Now consider a problem encountered in this thesis. The sample-window interface velocity history $u_0(t)$ is measured experimentally. The goal is to infer the flow history within the sample. Let the sample occupy the half-space $X \leq 0$ and the window occupy the half space $X \geq 0$. The boundary condition available is $u(0, t) = u_0(t)$.

Assume $u_0(t)$ is zero until some instant $t_0 > 0$. The $C_-$ characteristics emanating from the positive $X$-axis carry with them $J_- = 0$, therefore the flow in the window region is simple. The measured velocity history $u_0(t)$ is the “input” for the problem in the window, the same role as $\sigma_{x_0}(t)$ in the previous problem. Assuming the
isentropic equation of state of the window is known, the measured velocity \( u_0(t) \) can be converted to the stress at the interface. Since the stress is continuous everywhere (where there are no shock waves), the stress at the interface on the sample side is obtained.

Now the problem is converted to the following: for a sample occupying the half-space \( X \leq 0 \), a time-dependent stress \( \sigma_{x0}(t) \) is applied at \( X = 0 \). What is the flow history in the sample? This problem seems to be the same as the problem in Section 2.3.1, but there is one crucial difference: the Riemann invariants are not continuous along the same characteristic across the interface. This is because there is a slight impedance mismatch between the two materials. Therefore, it is necessary to modify the scheme. To minimize this mismatch, it is desirable to choose a window with similar impedance as the sample. The impedance is defined to be \( \rho_0 c_L \), which describes a material’s “stiffness” or its resistance to shock compression.

For the \( C_+ \) characteristics in the \( X > 0 \) (window) region, it is still true that \( J_+^w = 2\phi^w[\sigma_{x0}(t)] = 2u_0(t) \). At the interface, \( J_+^s + J_-^s = 2u_0 = J_+^w \), leading to \( J_+^s = J_+^w - J_-^s = (u_0 + \phi^w) - (u_0 - \phi^s) = \phi^w + \phi^s \). The algorithm is now the following. Use the EOS of the window and the sample to find the values of \( \sigma_{x0}(t) \) and \( \phi^s(t) \). Compute the values of the Riemann invariants in the sample region: \( J_+^s = \phi^w + \phi^s \), \( J_-^s = \phi^w - \phi^s \). The values of \( u \) and \( \phi \) at the intersection \((i, j)\) can be computed by \( u(i, j) = (J_+^s(i) + J_-^s(j))/2 \), \( \phi(i, j) = (J_+^s(i) - J_-^s(j))/2 \). The values of \( c_L, \sigma_x \) can be obtained from \( \phi \). The location of the intersection \((i, j)\) can be determined by a similar method as in Section 2.3.1.

There are other types of problems with different variants of the method, but they are beyond the scope of this thesis.
2.4 Geometry of Crystals

The properties of a macroscopic body is, in large part, determined by how its atoms are arranged. A crystal is composed of atoms arranged in a pattern that is periodic in three dimensions. At finite temperatures, atoms in a crystal are always in motion, but the equilibrium positions of the constituent atoms still form a periodic pattern.

The physical properties of a crystal are determined by the underlying periodic pattern and its symmetry. For example, at ambient conditions, carbon exists as two different types of crystals, diamond and graphite, exhibiting radically different physical properties. Diamond is the hardest known natural material, has high yield strength, thermal conductivity, and is transparent to visible lights, while graphite is quite soft, has high electrical conductivity, and is opaque to visible lights.

When studying the behavior of matter at extreme conditions, it is important to characterize its crystalline structure before, during and after compression. Experimentally, the technique of x-ray diffraction is often used to probe the crystal structures because the wavelength of x-rays is comparable to the interatomic distances of atoms in crystals. This will be discussed in Section 2.5. In this section, the geometry of crystals is summarized.

To describe the translational symmetry of a crystal it is useful to abstract the building blocks of the periodic pattern as points. If a crystal is invariant under translations by vectors \( \mathbf{a}, \mathbf{b}, \mathbf{c} \) (called primitive vectors), then the crystal can be represented by a three-dimensional Bravais lattice consisting of all points of the form \( \mathbf{R} = n_1 \mathbf{a} + n_2 \mathbf{b} + n_3 \mathbf{c} \), where \( n_1, n_2, n_3 \) are integers. The unit cell of the Bravais lattice is defined to be the three-dimensional volume spanned by the three primitive vectors. Each unit cell effectively possesses one and only one point. The lengths \( a, b, c \) of the primitive vectors and the angles \( \alpha, \beta, \gamma \) between them are called lattice parameters of the unit cell. Specifying different values to the lattice parameters will produce unit cells of various shapes and therefore various kinds of point lattices.
Figure 2.4: A point lattice, where each unit cell represents a building block of the periodic pattern of a crystal. A unit cell is highlighted and the primitive vectors $\mathbf{a}, \mathbf{b}, \mathbf{c}$ and lattice parameters $a, b, c, \alpha, \beta, \gamma$ are shown.

The space group of a Bravais lattice is the set of symmetry operations (such as translation, and rotation, and a combination of both) of the lattice. The subset of the space group consisting of only rotational symmetries is called the point group of the Bravais lattice. There are only 7 distinct point groups, corresponding to seven crystal systems: cubic, tetragonal, orthorhombic, monoclinic, triclinic, trigonal, hexagonal. There are 14 distinct space groups, corresponding to 14 Bravais lattices. The cubic point group contains 3 space subgroups: simple cubic (sc), body-centered cubic (bcc) and face-centered cubic (fcc). The hexagonal point group contains just one space subgroup: the simple hexagonal (sh). If the asymmetry of the building blocks themselves are considered (for example, a building block consists of two atoms, so the building block itself has a preferred direction, thus is not full spherically symmetric), then the 7 point groups split into 32 crystallographic point groups, and the 14 Bravais space groups split into 230 crystallographic space groups. They are too numerous to enumerate.

Each point of the Bravais lattice is an abstraction of the building blocks of the
periodic pattern of a real crystal. In the simplest situation, the building block is a single atom. It is also possible that the building block consist of two or more atoms, of the same kind or different kind. These are described by a lattice with a basis, each basis being a vector of fractional linear combination of the primitive vectors. For example, the cubic diamond lattice is an fcc lattice with a basis having two atoms, located at $(0,0,0)$ and $(1/4,1/4,1/4)$. The hexagonal close-packed (hcp) lattice, the structure of titanium, is a sh lattice with a basis of two atoms located at $(1/3,2/3,1/4)$ and $(2/3,1/3,3/4)$.

If the material consists of only one kind of atom, it is always possible to find a primitive cell containing only one atom. The most common such choice is the Wigner-Seitz cell, defined to be the three-dimensional volume closer to a lattice point than any other lattice points. However, a primitive cell is usually not the most convenient or visually illuminating unit cell to use. If the underlying symmetry is cubic or hexagonal, conventional unit cells are often used. The conventional unit cell is generally chosen to be bigger than the primitive cell. For example, a bcc lattice can be described by primitive vectors, but it can also be described by a cubic unit cell with a basis, $(0,0,0)$ and $(1/2,1/2,1/2)$. Similarly, a fcc cell can be described by a cubic unit cell with a basis, consisting of four atoms located at $(0,0,0)$, $(1/2,1/2,0)$, $(1/2,0,1/2)$, $(0,1/2,1/2)$.

In solid state physics, the concept of reciprocal lattice is important for describing the scattering processes, such as x-ray diffraction. The reciprocal lattice is defined to be set of points $\mathbf{K}$ in the wave vector space that satisfies the relation

$$e^{i\mathbf{K} \cdot \mathbf{R}} = 1$$

(2.26)

for all $\mathbf{R}$ in the Bravais lattice. The reciprocal lattice itself is a Bravais lattice, with
the primitive vectors

\[ a^* = 2\pi \frac{b \times c}{a \cdot b \times c}, \quad b^* = 2\pi \frac{c \times a}{a \cdot b \times c}, \quad c^* = 2\pi \frac{a \times b}{a \cdot b \times c}. \]  

(2.27)

The vector \( a^* \) is perpendicular to \( b \) and \( c \), and \( a^* \cdot a = 2\pi \). Similar properties hold for the other primitive vectors.

It is possible to partition a three-dimensional Bravais lattice into a family of parallel, equally spaced lattice planes, which together contain all the points of the lattice. Each reciprocal lattice vector \( K \) corresponds to a family of lattice planes perpendicular to \( K \) and separated by a distance \( d \), where \( 2\pi/d \) is the length of the shortest reciprocal lattice vector parallel to \( K \). Conversely, for any family of lattice planes separated by a distance \( d \), there exist reciprocal lattice vectors perpendicular to the family of planes, the shortest of which have a length of \( 2\pi/d \). Hence the normal direction of each family of lattice planes corresponds to a direction in the reciprocal space. Let \( K \) be the shortest reciprocal vector in this direction, written as \( K = h\mathbf{a}^* + k\mathbf{b}^* + l\mathbf{c}^* \). The integers \( h, k, l \) are called the Miller indices of the family of lattice planes. The family of lattice planes is usually written as \( (hkl) \). The symbol can also represent a direction in the reciprocal lattice. \( \langle hkl \rangle \) refers to the set of all families of lattice planes related to \( (hkl) \) by rotational symmetry. For example, in a simple cubic lattice, the families of lattice planes \((100), (\overline{1}00), (010), (0\overline{1}0), (001), (00\overline{1})\) are collectively written as \( \langle 100 \rangle \).

### 2.5 X-Ray Diffraction

X-ray diffraction (XRD) is an important tool to investigate the microscopic structure of matter. The wavelengths of x-rays are comparable typical interatomic distances of crystals, so x-rays scatter off crystals in the way that ordinary light scatters off a diffraction grating. By measuring the intensity of diffracted x-rays at different
Figure 2.5: The geometry of diffraction by a monochromatic planar x-ray. $\mathbf{k}_0$ is the wave vector of the incident monochromatic plane x-ray. The x-ray is diffracted by a crystal whose atoms are located at $\mathbf{R}_l$ ($l$ indexes the atoms), and the observational device is located far away from the crystal, in the $\mathbf{r}$ direction. The x-rays are scattered into all directions, but the intensity is not spherically symmetric, and depends on $\mathbf{r}$. Equation (2.28) describes the intensity of x-rays scattered into $\mathbf{r}$ direction relative to the intensity of the incident x-ray.

directions, it is possible to infer properties of the crystals, such as their constituents and lattice parameters. In this sense, x-rays are probes converting microscopic properties into macroscopic observables.

The interaction of x-rays with matter is quite complicated. At a reduced level, it is sensible to start from the interaction of x-rays with an atom. The incident x-ray exerts a periodic force on the charged components of the atom, such that the particles oscillate at the frequency of the incoming x-ray and emit x-rays of the same frequency at all directions. The direction-dependent intensity of scattered x-rays by an atom is an property of the atom and the polarization of the incoming x-ray, and is recorded as the atomic scattering factor $f(\mathbf{r})$. The crystal consists of a large collection of atoms, each contributing to the scattering of the incoming x-ray. If the crystal consists of atoms located at $\mathbf{R}_l$ for the $l$th atom, the relative intensity of scattered x-ray per solid angle far away from the crystal is

$$I(\mathbf{r}, t) = \left| \sum_l f_l(\mathbf{r}, t) e^{i \mathbf{q} \cdot \mathbf{R}_l} \right|^2,$$  \hspace{1cm} (2.28)
where \( \mathbf{q} = \mathbf{k}_0 - \mathbf{k}, \mathbf{k}_0 \) is the incident wave vector, \( \mathbf{k} = k_0 \hat{r} \) is the outgoing wave vector. The angle \( 2\theta \) between \( \mathbf{k}_0 \) and \( \mathbf{k} \) is the scattering angle. In the context of x-ray diffraction, \( \theta \) is usually called the Bragg angle.

### 2.5.1 Diffraction from a Bravais Lattice

If the crystal consists of the same type of atoms, then

\[
I(\hat{r}, t) = |f(\hat{r}, t)|^2 \left| \sum_l e^{i\mathbf{q} \cdot \mathbf{R}_l} \right|^2.
\]

(2.29)

The diffraction condition is that \( \mathbf{q} \cdot \mathbf{R} \) is a multiple of \( 2\pi \) for every \( \mathbf{R} \) in the Bravais lattice. That is to say, the vector \( \mathbf{q} \) belongs to the reciprocal lattice. If this condition is satisfied, then all terms in the sum of Equation (2.29) are 1, so the intensity takes its maximum. The diffraction condition dictates the change \( \mathbf{q} \) of wave vectors, so diffraction only occurs at a few selected directions.

If \( \mathbf{q} \) is a reciprocal vector satisfying the diffraction condition, then it can be written as \( \mathbf{q} = n\mathbf{K} = nha^* + nk\mathbf{b}^* + nl\mathbf{c}^* \), where \( \mathbf{K} \) is the shortest reciprocal vector parallel to \( \mathbf{q} \), and the integer \( n \) is the order of diffraction. The length of \( \mathbf{K} \) is \( K = 2\pi/d \), where \( d \) is the distance between the family of lattice planes perpendicular to \( \mathbf{q} \). Combining with the relation \( \mathbf{q} = \mathbf{k}_0 - \mathbf{k} = \mathbf{k}_0 - k_0 \hat{r} \), the diffraction condition can be rewritten as

\[
2d \sin \theta = n\lambda.
\]

(2.30)

This is Bragg’s law. The distance \( d \) is sometimes written as \( d_{hkl} \), where \( hkl \) are Miller indices of the vector \( \mathbf{q} \), to emphasize the direction of \( \mathbf{q} \).

As an example, consider the simple cubic lattice whose lattice parameter is \( a \). Its reciprocal lattice is again a simple cubic whose lattice parameter is \( 2\pi/a \). The allowed \( \mathbf{q} \) are points of the reciprocal lattice, corresponding to the families of lattice planes \( \langle 100 \rangle, \langle 110 \rangle, \langle 111 \rangle, \langle 200 \rangle, \langle 210 \rangle, \langle 211 \rangle, \langle 220 \rangle, \langle 221 \rangle, \langle 300 \rangle \), etc.
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2.5.2 Diffraction from a Lattice with a Basis

If the crystal is a lattice with a basis, then the position of every atom in the crystal can be written as \( \mathbf{R} = \mathbf{u}_j + \mathbf{v}_l \) for some \( j \) and \( l \), where \( \mathbf{u}_j \) is a vector of the Bravais lattice, and \( \mathbf{v}_l \) is a vector of the basis. The relative intensity per solid angle is then

\[
I(\hat{r}, t) = \left| \sum_l f_l(\hat{r}, t) e^{i\mathbf{q} \cdot \mathbf{v}_l} \right|^2 \left| \sum_j e^{i\mathbf{q} \cdot \mathbf{u}_j} \right|^2 = |S_\mathbf{q}|^2 \left| \sum_j e^{i\mathbf{q} \cdot \mathbf{u}_j} \right|^2
\]

(2.31)

The second factor gives the diffraction condition as before. At each direction \( \mathbf{q} \) allowed by the diffraction condition, the geometrical structure factor \( S_\mathbf{q} \) further modifies the intensity of the diffracted beam. It is even possible that \( S_\mathbf{q} \) is zero for some directions \( \mathbf{q} \), such that the diffraction peak corresponding to \( \mathbf{q} \) vanishes. This is caused purely by the existence of the basis in the lattice.

As an example, the bcc lattice whose conventional unit cell has lattice parameter \( a \) can be regarded as a simple cubic lattice with a basis \( (0, 0, 0), (\frac{1}{2}, \frac{1}{2}, \frac{1}{2}) \). Since \( \mathbf{q} = n\mathbf{a}^* + n\mathbf{b}^* + n\mathbf{c}^*, \mathbf{v}_1 = 0, \mathbf{v}_2 = (\mathbf{a} + \mathbf{b} + \mathbf{c})/2 \), the geometrical structure factor is \( S_\mathbf{q} = f + f e^{i\pi n(h+k+l)} \), which is zero if and only if \( n(h+k+l) \) is odd. Therefore, some of the allowed directions discussed in the last subsection can cause \( S_\mathbf{q} \) to vanish. For example, the \( \langle 100 \rangle, \langle 111 \rangle, \langle 210 \rangle, \langle 221 \rangle, \langle 300 \rangle \) directions, although allowed by the diffraction condition, produce zero intensity. This phenomenon, called systematic absence, can be useful to distinguish different structures of the same Bravais lattice having a different basis.

2.5.3 Single Crystal and Powder Diffraction

The diffraction condition assumes a monochromatic plane x-ray diffracts from a single crystal. This condition is in practice quite hard to satisfy, rendering it useless. There are two ways to overcome this: the Laue method (single crystal method) and Debye-Scherrer method (powder diffraction method).
The Laue method still uses a piece of single crystal but allows the x-ray to be non-monochromatic. By making the spread in wavelengths sufficiently large, one can be sure of finding some reciprocal lattice points satisfied by a certain wavelength. The Laue method is best suited for determining the orientation of a single crystal whose structure is known.

The Debye-Scherrer method still uses monochromatic x-rays, but allows the crystal to rotate. In practice, this is achieved by using a polycrystalline sample or a powder sample, grains of which are large enough to produce coherent x-ray scatterings. The grains are randomly oriented, so the resulting diffraction pattern is the same as that produced by all possible orientations of a single crystal. Each reciprocal lattice vector rotates around the incident direction to generate a cone of scattered x-rays. The angle between the scattered x-ray and the axis of the cone is the scattering angle $2\theta$. If the sample is completely randomly oriented, the intensity of the scattered x-rays is uniform around a cone. In reality, some polycrystalline samples have a preferred direction, so the orientation is not completely random. In this case, the intensity around the cone is instead azimuthally dependent and reveals details about the “texture” of the sample. The Debye-Scherrer method is suited for determining lattice parameters with high precision and for identification of phases. Further discussions of x-ray diffraction can be found in [32].
Chapter 3

Experimental Methods

3.1 Structure Measurement Using PXRDIP

At OMEGA and OMEGA-EP, x-ray diffraction is achieved via a diagnostic called Powder X-Ray Diffraction Image Plates (PXRDIP) \cite{33}. A similar diagnostic called TARget Diffraction In Situ (TARDIS) has been developed and implemented on the National Ignition Facility (NIF) \cite{34}. The diagnostic uses the Debye-Scherrer method, where a polycrystalline sample or a powder is irradiated by a monochromatic x-ray beam. Typically, the grains of the sample are microscopically large enough to produce detectable diffraction signals. Macroscopically, the grains are small and randomly oriented (possibly with a preferred direction), so the diffracted x-rays are a series of coaxial cones. Each cone corresponds to a particular set of lattice planes $hkl$, and the angle between a cone ray and the axis is the scattering angle $2\theta$ for the family of lattice planes $hkl$.

The diagnostic hardware consists of a $50 \times 50 \times 75 \text{ mm}^3$ rectangular with a $10 \times 10 \text{ mm}^2$ cavity on the front square plate, where the target assembly is mounted, and a $25 \text{ mm}$ diameter circular aperture on the back square plate, allowing VISAR probe beam and the self-emission of the target to pass through. The side walls and the back plate of the PXRDIP box are $1\text{ mm}$ thick stainless steel, while the front plate is $2 \text{ mm}$ tantalum for additional x-ray shielding.

The diffracted x-rays are recorded by image plates lining the inside of the PXRDIP
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Figure 3.1: The PXRDIP diagnostic and target design. In a typical x-ray diffraction experiment using PXRDIP, the sample is sandwiched between an ablator and a transparent window, and then mounted on top of a pinhole substrate. The target package is installed on the front plate of PXRDIP. The target is compressed by irradiating a laser beam on the ablator. The x-rays are generated by irradiating a foil with another laser pulse. The diffracted x-rays are recorded by the image plates in the PXRDIP box. On the backside, an opening allows the passage of VISAR beam and the self-emission of the target to pass through.

The image plates are covered by a 25 µm-thick black Kapton sheets and a 13 to 100 µm thick metal foil to filter out background x-rays other than the wavelength used for diffraction.

The x-rays are generated by irradiating a metal foil with a 1ns laser pulse. The laser irradiance is chosen to optimize x-ray conversion efficiency of Cu He-α (1.4816 Å) or Ge He-α (1.2097 Å) [35].

The Debye-Scherrer cones intersect the image plates to produce a series rings. The image plates are projected onto the $2\theta - \phi$ plane, where $2\theta$ is the scattering angle and $\phi$ is the azimuthal angle around the direct x-ray direction. In this plane, Debye-Scherrer rings are projected into constant $2\theta$ lines. A statistic-sensitive nonlinear
CHAPTER 3. EXPERIMENTAL METHODS

iterative peak-clipping algorithm is used to estimate and subtract the spatially slowly varying background. Other features not originating from the compressed sample or pinhole substrate are masked out when taking the lineout of images. The pinhole diffraction peaks are used for geometry calibration to accurately locate the diffraction scattering angles $2\theta$. A systematic correction to $2\theta$ is made to account for the fact that the pinhole substrate is slightly displaced from the sample.

3.2 Velocity Measurement Using VISAR

The determination of stress in the sample using the method of characteristics requires the measurement of the sample-window interface velocity. This is achieved using a Line-imaging Velocity Interferometer System for Any Reflector (VISAR) [36]. A review of VISAR is found at [37, 38].

VISAR is based on the measurement of the Doppler shift in an optical probe laser beam reflected from a moving reflecting surface (such as a shock front, a free surface, or an interface). Detection of the Doppler shift is performed using interferometric techniques combined with high speed detectors such as streak cameras. A glass delay element called etalon is installed in one of the paths, causing a time delay $\tau = (2h/c)(n - 1/n)$, where $h$ is the thickness of the etalon, and $n$ its refractive index. The phase difference between the two legs is $\phi = 2\pi c\tau / \lambda$. The probe beam reflecting off the moving measure surface is subjected to Doppler shift, and the change of its wavelength is proportional to the velocity of the moving surface, $\Delta \lambda = -(2\lambda/c)u(t)$. The change of the phase difference is also proportional to the velocity, $\Delta \phi = -2\pi c\tau \Delta \lambda / \lambda^2 = (4\pi \tau / \lambda)u$. Therefore, the velocity of the moving surface can be obtained by accurately measuring the change in the phase difference via counting fringe shifts.

On OMEGA EP, the reflected VISAR probe beam of 532 nm together with the self-emission of the target is collected by an optical relay, and passes through a dichroic
mirror, which reflects the 532 nm VISAR probe beam into VISAR. The remaining light is allowed to pass through and is collected by the SOP system discussed in Section 3.3. The light reflected into VISAR is projected through an entrance slit into a narrow linear region, which is streaked in time by the streak camera, yielding a two-dimensional image.

If the moving surface is a shock or an interface, the VISAR probe beam travels through a transparent window, and the refractive index of the window changes how fast the surface appears to move, just like how a swimming pool appears shallower than it actually is. The measured velocity is called apparent velocity and denoted \( u_a \), while the true velocity is denoted \( u_t \). The window is compressed during the experiment, causing its refractive index to change, further complicating the relation between \( u_a \) and \( u_t \). If the compression wave into the window is simple, no shocks occur in the window, and the refractive index of the window varies linearly with density \( n = a + b\rho \), then the true velocity is related to the apparent velocity by \( u_t = u_a/a \) [39]. The coefficients \( a \) and \( b \) are properties of the window material, and can be measured experimentally. Kirsch et al. [40] has measured a more complicated, nonlinear relation between \( u_a \) and \( u_t \) for LiF.

### 3.2.1 Velocity Analysis using Fourier Transform Method

The VISAR image is analyzed to extract fringe phase as a function of space and time. The most commonly used method in the HED community is the Fourier transform method (FTM). The method assumes that the recorded fringe intensity as a function of time \( t \) and position \( x \) is represented as

\[
S(t, x) = B(t, x) + A(t, y) \sin[\delta(t, x) + 2\pi f_0 x + \delta_0],
\]

(3.1)

where \( B(t, x) \) is the background intensity, \( A(t, x) \) is the fringe amplitude, \( 2\pi f_0 x + \delta_0 \) is the linear phase ramp (carrier wave) across the image along spatial direction,
and $\theta(t, x)$ is the phase modulation superimposed on the carrier wave containing the velocity information. The functions $A(t, x)$ and $B(t, x)$ need to vary slowly both spatially and temporally, and $\phi(t, x)$ needs to vary slowly spatially for this method to work. If these conditions are satisfied, then at each instant $S(t, x)$ is a sinusoidal function of $x$.

In real data, the fringe pattern is rarely composed of only one frequency, as shown in Figure 3.2a. One can perform a Fourier transform in spatial direction to examine the most prominent frequencies. In most cases, the power spectra attains its maximum at the zero frequency; this corresponds to the slowly varying background $B(t, x)$, or the “DC” component. There are usually two extra peaks, at equal distances to the zero frequency peak. The absolute value of the frequencies of these peaks are selected as the carrier wave frequency $f_0$. The fringe pattern is mostly due to oscillations with frequencies close to $f_0$. In Figure 3.2b, the absolute value of the Fourier coefficients of the data in (a) is shown in the form of a spectrogram, along with its lineout in the spatial direction. The Fourier coefficient peaks at three locations: 0 and $\pm f_0$. One can use a filter window to pick out frequency components near $f_0$, and perform an inverse Fourier transform to recover a “purer” fringe pattern $Z(t, x)$, without low-frequency, slowly varying components, such as those caused by nonuniform illumination, and high-frequency, rapidly varying components, likely due to streak camera noises. The complex function $Z(x, t)$ is

One then extract the phase $\theta(t, x) + 2\pi f_0 x + \delta_0$ from the complex function $Z(x, t)$ by evaluating its argument $\arg Z(x, t)$ and fringe amplitude by evaluating its module $|Z(t, x)|$. Figure 3.2c shows the extracted phase. In practice, the arctan function in most computer languages yields values only between $-\pi$ and $\pi$. The lineout of phase at a given instant has a saw-tooth form, as shown in Figure 3.2c. At each point in space, a multiple of $2\pi$ can be added to the phase to turn the saw-tooth form into a linear ramp. This operation is called “unwrapping” in the spatial direction, and the result is shown in Figure 3.2d. At different instants, the spatial phase are parallel
Figure 3.2: Main steps of phase extraction using the Fourier Transform Method. (a) a typical VISAR image from a ramp compressed target. (b) The Fourier coefficients of the data in (a) shown as a spectrogram, along with its lineout, showing three peaks $0$ and $\pm f_0$, the carrier frequency. (c) The extracted phase $\theta(t, x) + 2\pi f_0 x + \delta_0$, constrained within $[-\pi, \pi]$. The right panel shows the phase lineout at $t = 0$, taking the form of a saw-tooth wave. (d) The spatially unwrapped phase. The lineout at each instant is a linear ramp, but the ramps at different instants may differ by a relative phase. The relative phase is shown as the red curve. (e) The temporally unwrapped phase, which is the true $\delta(t, x) + 2\pi f_0 x + \delta_0$. The relative phase is more or less continuous, consistent with visual observation of (a). (f) The map of $\delta(t, x)$, which is almost constant in $x$ and varies in $t$. It is converted to velocity by $u(t) = \theta(t, x) \cdot \text{VPF}/(2\pi)$. 
linear ramps displaced from each other. The displacement, or the relative phase is shown as the red curve. A multiple of $2\pi$ can be added to the lineout to produce a more continuous relative phase, resulting in Figure 3.2e. This operation is called “unwrapping” in the temporal direction. The result is the true $\theta(t, x) + 2\pi f_0 x + \delta_0$ without being restricted between $-\pi$ an $\pi$. By removing the linear ramp $2\pi f_0 x + \delta_0$, one obtains the information-containing phase function $\theta(t, x)$. The phase function can be converted to velocity via

$$u(t) = \frac{\dot{\theta}(t, x)}{2\pi} \cdot \text{VPF.} \quad (3.2)$$

where the velocity per fringe (VPF) is the sensitivity calibration of the VISAR setup.

### 3.2.2 Ghost Fringe Subtraction

Often experimental VISAR images contain ghost fringes, which are extraneous fringes present across the entire image, parallel to the time axis. They result from a reflection off a nonmoving surface, and can contaminate the true VISAR signal. In Figure 3.3, the VISAR image of channel 2 from OMEGA EP shot 25879 is shown. The ghost fringes are particularly visible at later times (>8 ns) when the moving fringe data is weak. The velocity extracted using the FTM method discussed in Section 3.2.2 results in the red curve, which is clearly not the true velocity. The ability of extracting velocity is compromised by the existence of the ghosts.

Ghost fringes arise when the experiment contains a partially reflecting surface different from the measurement surface, usually a vacuum interface that creates a ghost reflection. In experiments where samples are tampered by a transparent window, the window back surface can act as such a surface. Antireflection coatings are usually applied to the back surface to mitigate the problem, but the quality of the coatings can vary. In the unfortunate situations that ghost fringes appear, there are a number of ways to remove them, none of which is perfect.
Figure 3.3: The VISAR image of channel 2 of shot 25879. The image is dominated by ghost fringes manifested as constant phase fringes at later times (>8 ns), although it is obvious by human eye that there are fringe movement, ramping up from 8 to 10 ns, and slowing down from 11 to 12 ns. The red curve is the velocity profile obtained by the Fourier analysis method, which clearly does not capture the fringe movement.
Figure 3.4: A typical target configuration from which ghost fringes may arise. The partially reflective back surface of the LiF window reflects a portion of the incoming VISAR probe beam, producing a background of static fringes that overlay the fringe pattern of the dynamically driven surface that is being probed by the experiment.

A popular method of distinguishing the ghost from science components is to take a Fourier transform, and hope that the science and ghost components have different frequencies. Another method [36] uses the fringe contrast as a measure of the magnitude of the ghost signal and develops analytic corrections to the fringe phase based on the contrast shift. The method described in this subsection is based on the method by D. J. Erskine et al. [41, 42].

When there exists a nonmoving free surface, a portion $G$ of VISAR probe beam is reflected before entering the window. Let $S_p(t)$ be any target incandescence or detector bias, $L(t)$ be the intensity of the VISAR probe beam (may be time dependent), $R(t)$ be the reflectance of the sample-window interface, $\kappa(t, s)$ be the absorption
coefficient of the window, $\gamma$ be the system contrast. With the aid of Figure 3.4, the VISAR intensity is

$$S(t, x) = S_b(t) + L(t)(1 - G)R(t)e^{-\int_{x(t,s)}^x ds}[1 + \gamma \sin[\phi(x) + \theta(t)]]$$

$$+ L(t)G[1 + \gamma \sin \phi(x)]$$

$$= [S_b(t) + L(t)G] + L(t)(1 - G)R(t)e^{-\int_{x(t,s)}^x ds} \quad \text{(background intensity)}$$

$$+ L(t)G\gamma \sin \phi(x) \quad \text{(ghost fringe)}$$

$$+ L(t)(1 - G)R(t)e^{-\int_{x(t,s)}^x ds} \gamma \sin[\phi(x) + \theta(t)], \quad \text{(science fringe)}$$

(3.3)

where $\phi(x) = 2\pi f_0 x + \delta_0$ is the linear phase ramp. Using trigonometric identities, the sum of ghost and science fringes can be written as

$$L(t)G\gamma \sin \phi(x) + L(t)(1 - G)R(t)e^{-\int_{x(t,s)}^x ds} \gamma \sin[\phi(x) + \theta(t)]$$

$$= [L(t)G\gamma + L(t)(1 - G)R(t)e^{-\int_{x(t,s)}^x ds} \gamma \cos \theta(t)] \sin \phi(y)$$

$$+ [L(t)(1 - G)R(t)e^{-\int_{x(t,s)}^x ds} \gamma \sin \theta(t)] \cos \phi(y)$$

$$= A(t) \sin[\phi(y) + \Delta(t)].$$

(3.4)

Compare this equation with Equation (3.1). If ghost fringes are present and one is not careful, one might think he/she has extracted the phase function $\theta(t)$ but in reality what he/she extracts is the modified phase function $\Delta(t)$, which depends on $\theta(t)$ and a number of other factors!

When $S_b(t)$ and $L(t)$ vary much slower than $R(t)$, there is roughly a linear relation between the background intensity and the science fringe amplitude. If the ghosts are removed correctly, we should obtain the linear relation. Let the science fringe amplitude be $F(t) = L(t)(1 - G)R(t)e^{-\int_{x(t,s)}^x ds} \gamma$, then $A(t) \sin \Delta(t) = F(t) \sin \theta(t), A(t) \cos \Delta(t) = L(t)G\gamma + F(t) \cos \theta(t)$. We are now motivated to represent the fringe amplitude and phase at time $t$ by a point in Cartesian coordinate
(\(A(t) \cos \Delta(t), A(t) \sin \Delta(t)\)). By allowing \(t\) to vary, a curve in the plane is obtained (Lissajous representation). The effect of ghost fringes is a constant addition to the \(x\)-component of the Lissajous points. If a vector \((G_x, G_y)\) is subtracted from all Lissajous points, another curve is obtained. \((G_x, G_y)\) is a good estimation of the ghost fringe contribution if the corrected amplitude \(F(t)\) and the background intensity satisfies the linear relation for all \(t\). The corrected Lissajous curve has amplitude

\[
F_c(t) = \sqrt{[A(t) \cos \Delta(t) - G_x]^2 + [A(t) \sin \Delta(t) - G_y]^2}.
\] (3.5)

and the background amplitude can be obtained as the zero frequency component of the Fourier coefficient. There are more sophisticated method to obtain the fringing and background amplitude, e.g. the speckle adaptive method by (again) D. J. Erksine [42].

In Figure 3.5, a solution of ghost fringe subtraction for s25879 VISAR image of channel 2 is presented. The corrected phase correctly captures the rise, plateau, and fall of the ramp compression wave, and is consistent with human eye observation.

A benefit of this analysis is the reflectance \(R(t)\) of the sample-window interface, because the fringe amplitude is proportional to \(R(t)\),

\[
R(t) = R(0) \frac{L(0) e^{-\int x(0,s) ds} F(t)}{L(t) e^{-\int x(t,s) ds} F(0)}.
\] (3.6)

If the VISAR probe beam intensity is kept constant, and the window absorption is ignored, then by obtaining the fringe amplitude from the ghost fringe subtraction process, \(R(t)\) can be readily obtained. The only unknown is \(R(0)\), the reflectance at ambient conditions, which can easily be estimated using the (hopefully) well-documented optical properties of window and sample materials at ambient conditions.
Figure 3.5: Ghost fringe subtraction solution of s25879 VISAR channel 2. (a) The background intensity as a function of time. (b) Corrected fringe amplitude $F_c(t)$. It is obtained choosing optimal vector $(G_x, G_y)$ and Equation (3.5). (c) The background intensity and the corrected fringe amplitude should satisfy a linear relation. The linearity is not perfect, especially at around 6 ns. (d) The corrected phase. Compared to the solution in Figure 3.3, the corrected solution correctly captures the rise, plateau, and fall of the ramp compression wave, consistent with human eye observation. In all figures, color represents time, where bluer (redder) colors represent earlier (later) times.
3.3 Temperature Measurement Using SOP

In dynamic compression experiments, a few temperature measurement techniques are available. The most common is Streaked Optical Pyrometer (SOP) which measures the self-emission of optical photons of a surface from the sample, usually an interface or a shock front, by assuming that the self-emission obeys Planck’s radiation law, which relates the spectral radiance (or summarily, the amount of radiation in a specified wavelength range) to temperature [43]. X-ray Absorption Fine-Structure (XAFS) spectroscopy [44], including X-ray Absorption Near Edge Structure (XANES) and Extended X-ray Absorption Fine Structure (EXAFS), measures the temperature from the Debye-Waller factor of the interference pattern when a photoelectron excited by incident x-rays is scattered by its neighbors. Because the XAFS methods rely on the interference effect, the materials to be probed are preferred to be solids.

Normally SOP does not lend itself to low temperature measurements due to low photon number emitted by a low temperature target over the detection duration. At laser facilities such as OMEGA and OMEGA EP, the typical detection duration is a couple of tens of nanoseconds. Still, it is desirable to use SOP for temperature measurement, even if only an upper bound, to obtain a constraint on the thermodynamic path of compressed samples, especially in experiments where geometry excludes the feasibility of XAFS methods. SOP is coupled to the VISAR, which is almost always fielded in experiments, so obtaining SOP data requires little extra effort.

In this section, SOP is introduced and the method of data analysis for high temperature samples is summarized. See [43] for a more detailed discussion and the calibration of the OMEGA SOP system. In Chapter 5, a modification of the method to cover low temperature regime is discussed.

Laser-driven dynamic compression experiments irradiate targets by lasers or by x-rays from a laser-driven hohlraum. The ablated surface is rapidly heated and expands outwards, generating compression waves into the target. A line-imaging VISAR
records the space and time resolved velocity history (with \( \sim 50 \) ps and \( \sim 10\text{–}20 \) \( \mu \)m resolution) of a shock front, a free surface, or an interface. VISAR also provide information about the surface’s reflectivity at the probe wavelength (532 nm). A heated shock front or interface emits blackbody radiation. The SOP simultaneously records the space-time history of the integrated spectral radiance of such blackbody radiation over 590 nm to 850 nm with similar temporal and spatial resolutions.

The line-imaging VISAR and SOP are coupled and almost always field together. Both the reflected VISAR probe beam and the self-emission from the rear side of the target are collected by an \( f/3.3 \) telescope and optically relayed towards a dichroic mirror, where the VISAR probe beam is reflected while the self-emission passes towards the SOP cabinet. The entrance to the SOP cabinet is a long-pass filter (OG590) that transmits only light with a wavelength greater than 590 nm, filtering out the higher harmonics of the OMEGA/OMEGA EP laser. Inside the cabinet, the self-emission is imaged onto the external slit of a Rochester Optical Streak System (ROSS) camera. Inside the ROSS camera, the self-emission is focused onto the S20 photocathode deposited on a sapphire window. Photoelectrons are accelerated through the streak tube toward the phosphor screen. Photons emitted by the phosphor screen are transported through a 1:1 fiber-optic taper that is bonded directly to the charge-coupled-device (CCD) sensor. The accelerated electron beams are deflected by a time-varying electric field to achieve temporal resolution.

The SOP camera output of a single pixel in analog-to-digital units (ADU’s) is given by

\[
\langle I \rangle = \Delta t GG' \int d\lambda \Phi_2(\lambda; T) T'_x(\lambda) \text{SR}(\lambda) .
\]  

(3.7)

\( \Delta t \) is the dwell time on a single pixel, i.e. the amount of time that a given “streak” spends at a single pixel. \( G \) and \( G' \) are the streak camera gain of photoelectrons to CCD electrons, and CCD electrons to ADU’s, respectively. \( T'_x(\lambda) \) is the transmission spectra of any inserted ND or bandpass filters introduced to the system, and \( \text{SR}(\lambda) \) is
the SOP’s spectral system response function. \( \Phi_S(\lambda; T) \) is the spectral radiant power from the light source that maps to a single detector pixel, given by

\[
\Phi_S(\lambda; T) = \Delta A_{\text{src}} \Delta \Omega S(\lambda),
\]

(3.8)

where \( \Delta A_{\text{src}} \) is the light source area that maps to a single pixel, and \( \Delta \Omega \) is the solid angle of the \( f/3.3 \) telescope, and \( L_S(\lambda) \) is the source spectral radiance. For a Planckian radiator,

\[
L_S(\lambda; T) = \frac{2hc^2}{\lambda^5} \frac{1}{\exp(hc/\lambda kT) - 1}.
\]

(3.9)

By approximating \( T_x(\lambda)SR(\lambda) \) as a delta function, \( T_x(\lambda)SR(\lambda) = \Gamma \delta(\lambda - \lambda_0) \), where \( \Gamma \) is computed by integrating \( T_x(\lambda)SR(\lambda) \), the integration in Equation (3.7) can be reduced to a much simpler analytic form,

\[
T = \frac{T_0}{\ln(1 + A/I)}
\]

(3.10)

where

\[
T_0 = \frac{hc}{\lambda_0 k}, \quad A = \frac{2hc^2 \Gamma G' \Delta t \Delta A_{\text{src}} \Delta \Omega}{\lambda_0^5}
\]

(3.11)

In practice, \( A \) and \( T_0 \) can be calibrated using a standard light source for a particular experimental setup so that the result calculated using the simplified equation matches best with the measured result. These coefficients have been calibrated for OMEGA SOP system by Gregor et al. [43]. A similar calibration has been made for OMEGA EP but is unpublished.
Chapter 4

Exploring Phase Diagram of Silicon

4.1 Motivation

Silicon (Si) is one of the most abundant elements on Earth, and has been studied extensively at extreme conditions. X-ray diffraction on isothermally-compressed Si at room temperature using diamond anvil cell (DAC) technique reveals seven different structural phases, and four more during decompression [45]. Starting at ambient conditions, Si has a diamond cubic type structure (cd, Si-I, space group $Fd\bar{3}m$) [46], transitioning to Si-II ($\beta$-tin type, body-centered tetragonal (bct) structure, space group $I4_1/amd$) [47], Si-XI (an orthorhombic structure, space group $Imma$) [48, 49], Si-V (simple hexagonal structure, or sh, space group $P6/mmm$) [50], Si-VI (another orthorhombic structure, space group $Cmce$) [51], Si-VII (a hexagonal close-packed structure, or hcp, space group $P6_3/mmc$) [50], and finally Si-X (a face-centered cubic structure, or fcc, space group $Fm\bar{3}m$) [52], which remains stable to at least 248 GPa [53]. Anzellini et al. made the most recent systematic study of the phase boundaries by compressing Si more hydrostatically using helium as a pressure-transmitting medium [45].

Dynamic compression (shock, multishock, and ramp compression) experiments uniaxially load the sample and then the sample relaxes towards a more hydrostatic compression with time. Shock experiments have explored a variety of phase transitions in Si to 54 GPa [54–60]. Such transitions are usually inferred from wave
splittings in the measured particle velocity data [54–57] due to violation of the stability condition, \( \partial^2 p/\partial \rho^2 > 0 \), on the shock Hugoniot near the phase boundary. More recently, \textit{in-situ} x-ray diffraction was used to study solid-solid phase transitions and shock-induced melting of Si [58–60]. McBride \textit{et al.} [58] observed a lowering of the cd-\( \beta \)tin-\textit{Imma} phase boundary under shock compression. Turneaure \textit{et al.} [60] observed that the sh structure coexists with the liquid along the melting curve above 30 GPa, before Si completely melts at around 33 GPa. Completely melted Si samples recrystallize into the hcp structure under reshock for shocked states below 36.7 GPa.

Recent first-principle simulations using density functional theory (DFT) also provide important insight into the high-pressure and high-temperature phase diagram of Si [17–19]. Li \textit{et al.} predict that at ambient temperature, the sh structure transforms into a double-hexagonal close-packed (dhcp, space group \( P6_3/mmc \)) structure at 33 GPa before it transforms into hcp structure at 41 GPa [17]. The predicted dhcp structure has not been observed by ambient temperature experiments [45]. Paul \textit{et al.} [18, 19] also predicts the dhcp structure, albeit only above ambient temperature, and the principal isentrope crosses the \textit{Cmce}-dhcp boundary at 22 GPa, the dhcp-fcc boundary at 55 GPa, after which the fcc structure is stable to 2.8 TPa. All such calculations were performed assuming hydrostatic compression.

Single-shock experiments access only a limited part of the solid Si phase space due to the large temperature increase and subsequent melting upon shock compression beyond 33 GPa [60]. In contrast, ramp compression, by gradually increasing the pressure load on the sample, can achieve high pressure states along a path bounded by the principal isentrope and the principal Hugoniot. The work described here used ramp compression techniques to explore the structural evolution with pressure of solid Si to several hundred GPa (several Mbar).
4.2 Experimental Details

The experiments were performed on the OMEGA EP laser at the Laboratory for Laser Energetics, University of Rochester. The experimental configuration uses the powder x-ray diffraction image plates (PXRDIP) diagnostic [33] (Figure 4.1). The laser beam has an incident angle of 22.5° with respect to the target normal and a 1100-µm-diameter spot size with a super-Gaussian profile produced by distributed phase plates. The Si sample is ⟨100⟩-oriented, single crystal, 10- to 22-µm-thick, and is sandwiched between plates of ⟨110⟩-oriented, single crystal, 20- to 30-µm-thick diamond ablator, and a ⟨100⟩-oriented, single crystal, 100- to 120-µm-thick LiF window. The pieces are held together by epoxy that is approximately 1- to 2-µm-thick. The target stack is mounted on a 75-µm-thick W, Ta or Pt plate with a 300- or 400-µm-diameter pinhole aperture.

Ramp compression is accomplished by ablating the diamond with a laser pulse which gradually increases with power over 10 or 20 ns (Figure 4.1), creating a ramped compression wave that propagates through the target assembly. The impedance difference between these materials causes the ramped compression wave to reverberate within the Si layer as it is compressed to high pressure. The complete Si-LiF interface velocity history is accurately recorded to constrain this complex compression path.

When the Si sample achieves its peak compression, a backlighter foil (Cu or Ge) is illuminated with an 1-ns laser pulse to generate the x-rays used for diffraction. The laser irradiance is chosen to optimize x-ray conversion efficiency of Cu He-α (1.4816 Å) and Ge He-α (1.2097 Å) [35]. The x-rays are collimated by the pinhole, and diffract from both the compressed Si sample and the edge of the pinhole. The diffracted x-rays are filtered by Cu (or Al) and black kapton sheets and are recorded by image plates lining the PXRDIP box. The pinhole is not compressed at the time of x-ray illumination and is used to calibrate the geometry of diffraction experiments. The ambient crystal structures of W, Ta, and Pt are bcc, bcc, and fcc, respectively.
A line-imaging velocity interferometer system for any reflector (VISAR) [36] detects the Doppler shifts of a 532-nm probe beam reflected off Si-LiF interface to measure the interface velocity as a function of time. The measured apparent velocity is corrected to account for the refractive index change of LiF due to compression [40]. The method of characteristics [61, 62] is used to determine the pressure distribution of the sample, with the interface velocity as the boundary condition. HYADES hydrodynamic simulations [63] are used to corroborate the results of the method of characteristics [12]. A Monte-Carlo (MC) algorithm is used to estimate the mean pressure and its standard deviation, accounting for the uncertainties of velocity due to VISAR record [36], LiF refractive index [40], sample layer thickness, Si initial density [60], LiF initial density [64], and LiF equation of state [40]. We also record the 1σ interval of the pressure distribution during the 1-ns x-ray exposure, averaged over the MC samples, to characterize pressure nonuniformity.

The x-ray image plates are projected onto 2θ − φ plane, where 2θ is the scattering angle and φ is the azimuthal angle around the direct x-ray direction (Figure 4.2). In this plane, Debye-Scherrer rings are projected into constant 2θ lines. A non-linear iterative peak-clipping (SNIP) algorithm is used to estimate and subtract the spatially slowly varying background [34]. Other features not originating from the compressed Si sample or pinhole substrate are masked out when taking the lineout of images (see supplementary material). The pinhole diffraction peaks are used for geometry calibration to accurately locate the diffraction scattering angles 2θ. A systematic correction to 2θ is made to account for the fact that the pinhole substrate is slightly displaced from the sample [34].

### 4.3 Data and Analysis

Two distinct x-ray diffraction patterns from Si are observed between 40 and 389 GPa. The first pattern is observed in three experiments at 40(2) GPa, 51(3) GPa and
Figure 4.1: (a) The PXRDP experimental platform and diffraction data for shot 29637. A Si sample sandwiched between diamond ablator and lithium fluoride window is compressed with a 10- or 20-ns ramp laser pulse. Near peak compression, 1-ns laser pulse illuminates a Cu or Ge foil to produce He-α x-rays, which diffract from the compressed sample and are recorded by image plates lining the PXRDP box. VISAR is used to record the Si-LiF interface velocity throughout the compression. (b) The delivered ramp laser pulse (red) and the 1-ns square laser pulse used to produce the x-ray source (purple) for shot 29637. (c) The VISAR streak image and the extracted apparent Si-LiF interface velocities for shot 29637. (d) Average Si pressure and its standard deviation for shot 29637 determined by the method of characteristics.
Figure 4.2: X-ray diffraction measurements of Si at (a) 51(3) GPa and (b) 389(11) GPa, respectively. Rectangular image plates are digitally projected onto a $2\theta - \phi$ plane, where $2\theta$ is the scattering angle and $\phi$ is the azimuthal angle around $2\theta = 0$. Features not originating from the compressed Si sample or pinhole are masked out (green shade) when taking the lineout of images (see supplementary materials). (c) An azimuthally averaged lineout (black) of the diffraction image in (a) is compared with simulated diffraction patterns of three candidate structures, whose ideal peak locations are marked by vertical lines. Ideal peak positions from the pinhole material used for image plate calibration are shaded by gray, and peaks from the sample are shaded in red. (d) Lineout of the diffraction image in (b) is compared with the simulated fcc structure. Peaks from the sample are shaded in blue.
93(7) GPa and is consistent with hexagonal symmetry. As an example, diffraction
data for Si at 51(3) GPa (Figs. 4.2(a) and (c)) is compared with simulated hcp, dhcp
and sh structures with best-fit lattice parameters. This experimental lineout agrees
well with hcp structure with $a_{\text{hcp}} = 2.490(5)$ Å, $c_{\text{hcp}} = 4.199(11)$ Å, and $\rho_{\text{hcp}} = 4.14(2)$ g/cm$^3$. The dhcp structure, predicted by theory [17–19], is another strong
contender, whose lattice parameters are fit to $a_{\text{dhcp}} = 2.481(5)$ Å, $c_{\text{dhcp}} = 8.49(2)$ Å,
and $\rho_{\text{dhcp}} = 4.12(2)$ g/cm$^3$. However, the (103) peak is absent across the entire
azimuth in our observation. Though somewhat textured, the (104) peak is observed,
despite being predicted to be of lower intensity than (103) peak in the simulation.
The texture of the (104) peak observed does not allow the absence of (103) peak
across the $\sim 300^\circ$ observable azimuth angle in these experiments. This provides
enough evidence against the existence of a dhcp structure at this condition. The
simulated pattern for sh, using the hcp lattice parameters, agrees well with the data
albeit with some minor differences. However, there are two arguments against the sh
structure. The first being that the unit cell of sh structure contains only one atom, as
opposed to two atoms in the case of hcp, which indicates that the density is only half
of what can be expected from the isentrope calculated using DFT [65] (Figure 4.3(b)).
The second argument is that the axial ratio $c/a$ is 1.687(5), almost twice as big as the
theoretical value of 0.942 at 29 GPa and 0 K [18]. The sh structure is not close-packed
like hcp or dhcp, and we expect its $c/a$ to be close to 1. These two arguments rule out
the sh as a candidate for the observed structure. We therefore interpret the structure
as hcp, with a weighted average $c/a = 1.6861(9)$.

The second pattern is observed between 153 and 389 GPa, in seven different shots.
Diffraction data for Si at 389(11) GPa (Figure 4.2(b) and (d)) show the characteristic
fcc (111) and (200) peaks. The expected location of the third peak (220) for fcc
calculated using the best-fit $a_{\text{fcc}} = 2.996(8)$ Å would be $2\theta = 88.747^\circ$, which partially
overlaps with the pinhole (013) peak (Figure 4.2(d)).
4.4 Discussion

The measured $d$-spacings are compared to values calculated with density along the DFT isentrope [65] (Figure 4.3(a)). The DFT calculation [18, 19] predicts the dhcp phase along the DFT isentrope above 22 GPa, transitioning to fcc above 55 GPa [18, 65] (Figure 4.4(a)). In contrast, our data show that the hcp stability region extends above the isentrope over a pressure from at least 40 to 93 GPa, with a transition to fcc between 93 and 153 GPa. The data also confirm that the fcc phase is stable to at least 389 GPa, in agreement with the DFT calculation [18, 19].

The observation of the hcp phase along the ramp compression path between 40 and 93 GPa corroborates the results of a shock compression experiment [60]. Their data (Figure 4.4(b)) suggest the existence of a sh-hcp-liquid triple point, and a direct phase boundary between the sh and hcp phases without the predicted dhcp phase inbetween. The onset of the sh-hcp transition in [60] is close to the phase space assigned to be hcp by our data (red shaded in Figure 4.4(b)). The DAC data at ambient temperature and our ramp data together suggest a positive slope for the hcp-fcc phase boundary.

We discovered a significant increase in stability of the hcp structure for dynamically compressed silicon at pressures and temperatures where dhcp or fcc phases are predicted. Two potential explanations for this, include (1) the hcp phase is energetically favored, or (2) the hcp phase is somehow stabilized by the dynamic compression techniques used in this work, such as the presence of deviatoric stresses. It has been observed that phase diagrams measured in dynamic compression experiments can differ from those from hydrostatic DAC experiments. For example, a lowering of the cd-$\beta$tin-Imma phase boundaries in Si has been reported in dynamic compression experiments [58]. Also, it was observed that diamond remains in its ambient phase (FC8) up to 2 TPa under ramp compression, despite being predicted to transform into BC8 at 1 TPa [13]. Finally, the dhcp phase was observed at 5-20 GPa near the melting
boundary in DAC experiments [66] but not in dynamic compression experiments [67].

The densities calculated using the measured lattice parameters are higher than those along the measured Hugoniot [2] as expected, but systematically lower than the DFT isentrope [65] and the SESAME 3810 isentrope (Figure 4.3(b)). Due to the elastic-plastic transition, first-order phase transitions of the constituent target materials, and limitations in pulse shaping capabilities, the compression path contains multiple weak shocks [68] that increase the sample temperature compared to the principal isentrope. Though temperature is not measured (although we will present an effort in Chapter 5), the DFT isentrope and melt curve give a reasonable constraint on it (Figure 4.4(b)). On the other hand, the SESAME and DFT isentropes are not guaranteed to be correct. This is evident from that the $T = 100$ K isotherm from the same DFT calculation [19], while correctly capturing the density jumps at cd-$\beta$tin and sh-hcp transitions, overpredicts density compared to the DAC data at ambient temperature [45, 50–53] by about 15%. The $T = 100$ K SESAME isotherm is closer to DAC measurement at pressures higher than 50 GPa, but does not capture the density jumps at lower pressures. The same inaccuracy can happen along isentropes as well.

In conclusion, powder x-ray diffraction was used to measure crystal structure of Si between 40 and 389 GPa along a thermodynamic path close to the principal isentrope. Along this path, the hcp structure is stable to at least 93 GPa, a much higher pressure than predicted by simulations, and the fcc phase is stable from 153 GPa to at least 389 GPa. Data show no evidence for the predicted dhcp phase. This is likely due to that the hcp phase is more energetically favored, or the nonhydrostaticity of the loading path. These observations show that Si exhibits interesting and unexpected behavior under ramp loading not captured by theoretical calculations, motivating improved DFT calculations for equilibrium phase diagrams, and Molecular Dynamics simulations for phase transition pathways and kinetics [69].
Figure 4.3: (a) Pressure dependence of the measured d-spacings. The structure between 40 and 93 GPa is interpreted as hcp, and that between 153 and 390 GPa as fcc. The solid lines represent d-spacings of hcp (red) and fcc (blue) structures calculated using the DFT isentrope [65]. (b) Density-pressure phase diagram data of Si. Data of this work are shown in circles, DAC data [45–53] as crosses, and shock data [58–60] as triangles. For this work, the 1σ uncertainty in the mean pressure and the 1σ interval of the pressure distribution at the time of exposure are shown with larger and smaller caps on the error bars. Various curves are shown: 100 K isotherm and principal isentrope from SESAME 3810 table, 100 K isotherm, principal Hugoniot [19] and principal isentrope [65] from DFT calculation, and measured Hugoniot [2]. Our data show lower densities than both DFT and SESAME isentropes.
The solid-solid phase boundaries, calculated using DFT [18, 19], are shown as dashed good agreement with this work. Phases are shown. The reshocked states at 35.8 GPa and 36.2 GPa are sh/hcp/liquid mixtures, and the reshocked state at 39.3 GPa is hcp/liquid mixture. These data show up to at least 93 GPa, with first evidence of fcc at 153 GPa. The phases predicted by DFT are shaded with their respective colors. Along the isentrope, theory predicts that to reduce the number of lines. (b) The pressure-temperature phase diagram. The solid-solid phase boundaries, calculated using DFT [18, 19], are shown as dashed curves. DFT isentrope [65], SESAME-3810 isentrope, DFT Hugoniot [18], and melt curve [18] are also shown. Data of this work fall between the theoretical principal isentrope and the melt curve, indicated by striped red and blue regions for observed hcp and fcc structures, respectively. Shock data and reshock data [60] of various phases are shown. The reshocked states at 35.8 GPa and 36.2 GPa are sh/hcp/liquid mixtures, and the reshocked state at 39.3 GPa is hcp/liquid mixture. These data show good agreement with this work.
Chapter 5

Low Temperature SOP Measurements

Temperature measurement in ramp compression experiments has been a longstanding unsolved problem and goal in the HED community, because the temperatures are so low and there are fundamental difficulties with measuring such low temperatures using nanosecond scale pyrometry. As a result, many authors choose to infer temperature using hydrodynamic simulations [4] or assuming a thermodynamic path [1, 12, 13] such as an isentrope. In this chapter, we lay a foundation towards lowering the lower bound of temperatures that can be measured by pyrometry.

5.1 The Traditional Analysis Method and Its Limitations

The SOP records signals, $I$, in units of ADU, and there is a one-to-one correspondence between $I$ and the temperature $T$ by the relation

$$T = \frac{T_0}{\ln\left(1 + \frac{A_0(1-R)}{\eta I}\right)}$$

(5.1)

The parameters $I_0$ and $A_0$ are calibration constants, and depend on the neutral-density (ND) filter used in the experiment. Over the years, the components of the SOP on OMEGA and OMEGA EP have been changed and upgraded, so $I_0$ and $A_0$ have changed with time. For OMEGA EP SOP, if no ND filter is used,
Figure 5.1: The $I - T$ curves for OMEGA EP SOP, with $R = 0$ (perfect blackbody assumption), 1000 pixels in the temporal direction, and sweep duration 17 ns. The SOP signal is linearly related to the average number of photoelectrons per pixel generated at the photocathode of the SOP excited by the thermal photons, and they are shown in the same vertical axis with different scales. The noise floor of the SOP streak camera $\sigma_r = 6.435$ ADU is shown as the black dashed line. The one-photoelectron-over-2500-pixels level is shown as the red dashed line. As the signal approaches these critical points, the validity of the traditional SOP analysis method becomes unclear.
then $A_0 = 288\,600$ ADU/ns and $T_0 = 1.909$ eV before September 2018 \footnote{The system response of OMEGA EP SOP before September 2018 is not absolutely calibrated, but using quartz as temperature standard, it is found that one can use the system response and $A_0$ of OMEGA SOP \cite{43} multiplied by 0.6 as the system response and $A_0$ of OMEGA EP SOP before September 2018.} and $A_0 = 150\,179$ ADU/ns and $T_0 = 1.990$ eV after September 2018 \footnote{Calibrated by M. K. Ginnane, but the system response is unpublished.}. $\eta$ is the sweep speed, which is the number of pixels in the temporal direction divided by the sweep duration (typically a few dozens of nanoseconds). $R$ is the reflectance of the surface that is emitting the thermal photons and can be approximated by the reflectivity at 532 nm obtained by analyzing VISAR image.

The $I - T$ curves for OMEGA EP are shown in Figure 5.1, with $R = 0$ (perfect blackbody assumption), 1000 pixels in the temporal direction, and sweep duration 17 ns. The SOP signal is linearly related to the average number of photoelectrons per pixel generated at the photocathode of the SOP excited by the thermal photons (see Section 5.3 for details), and they are shown in the same vertical axis with different scales. Also shown is the noise floor of the SOP streak camera $\sigma_r = 6.435$ ADU (which will be calibrated in Section 5.5). As temperature decreases, $I$ also decreases until it comes close to the noise floor at 3970 K and 3380 K for the old and new OMEGA EP SOP, respectively. Below these temperatures, the signals are so low that they are almost buried in the detector noises.

When the signal level is low, not every detector pixel receives photoelectrons (or equivalently, thermal photons from the target). In the extreme case of $T = 0$, no pixels will receive photoelectrons at all. At finite but very low temperatures, the more detector pixels there are (while keeping the pixel size fixed, thus a larger solid angle), the more likely at least some pixels receive photoelectrons. In our applications, the region of interest on the SOP image is 20 pixels in the temporal direction and 125 pixels in the spatial direction, so totally 2500 pixels are used. The temperature at which 2500 pixels receive one photoelectron on average is roughly 2000 K for both SOP setup, much lower than the temperatures corresponding to the detector noise.
floors (see the red dashed horizontal line in Figure 5.1). In this case, the major factor challenging the validity of the traditional analysis method is the noise floor. If fewer pixels are used for evaluating the temperature (for example, if better temporal resolution or spatial resolution is desired), then the temperature corresponding to one electron over the region of interest will increase accordingly.

Assuming the classic propagation of uncertainty formula is applicable here, the relative uncertainty of $T$ is

$$\frac{\sigma_T^2}{T^2} = \frac{\sigma_{T_0}^2}{T_0^2} + \left( \frac{T}{T_0} \right)^2 \left[ 1 - \exp\left( -\frac{T_0}{T} \right) \right]^2 \left( \frac{\sigma_{A_0}^2}{A_0^2} + \frac{\sigma_{\eta}^2}{\eta^2} + \frac{\sigma_{1-R}^2}{(1-R)^2} + \frac{\sigma_{I}^2}{I^2} \right). \quad (5.2)$$

This inherently assumes that all quantities are uncorrelated. The relative uncertainties of various quantities are $T_0$ (0.2%), $A_0$ (3.5%), $\eta$ (1.5%), $(1 - R)$ (20%). The relative uncertainty of $I$ is usually 2% at very high temperatures (several eV) [43, 70].

For the SOP image shown in Figure 5.3, the relative uncertainty of $I$, averaged over regions of 20 × 125 pixels at the same spatial locations as the boxes, is shown in Figure 5.2. The formula is $\sigma_I/I = \sigma_I/(\sqrt{2500I})$. The relative intensity blows up at low intensities, takes a minimum at around 150 ADU, and stabilizes around 1.5% asymptotically.

The prefactor $x(1 - \exp(-x^{-1}))$ where $x = T/T_0$ in front of the second term of Equation (5.2) is shown in Figure 5.2(b). At $T = 5000$ K, $x = T/T_0 \approx 0.2$ for the new OMEGA EP SOP, so the value of the prefactor is roughly 20%. Therefore the relative uncertainty of $T$ is dominated by the second term of Equation (5.2) even at low temperatures. If the relative uncertainty of $I$ is indeed several thousands percent at low $I$ as shown in Figure 5.2(a), then the most crucial task of obtaining the relative uncertainty of $T$ at low temperatures is to estimate the relative uncertainty of $I$. Naively assigning a relative uncertainty of $I$ of several thousands percent can lead to a relative uncertainty of $T$ greater than one, meaning that the confidence interval of $T$ contains some negative values, which is absurd from a physics point of view.
Figure 5.2: (a) The relative uncertainty of SOP intensity $I$ averaged over $20 \times 125$ pixels at the same spatial locations as the boxes in Figure 5.3 for shot 32081. The formula is $\sigma_I/I = \sigma_I/(\sqrt{2500I})$. The relative intensity blows up at low intensities, and stabilizes around 1.5% asymptotically. (b) The prefactor $x(1 - \exp(-x^{-1}))$ where $x = T/T_0$ in front of the second term in Equation (5.2). At $T = 5000$ K, $x = T/T_0 \approx 0.2$ for the new OMEGA EP SOP, so the value of the prefactor is roughly 20%.

As far as I know, no author has studied the problem of how to estimate the uncertainty of $I$ at low temperatures. In this chapter, we lay the foundation for a framework for this task. It is apparent from the discussions above that this is related to the problem of assigning confidence intervals to a physical quantity ($T$ in this problem) when the domain of the quantity is constrained by the physics considerations (for example, $T$ must be positive).

5.2 A Look at a Typical SOP Image

In science, the usual way of measuring a quantity $q$ is to repeatedly measure it to obtain a sample $\{q_1, q_2, \ldots, q_n\}$, and compute the mean $\bar{q} = \sum_{i=1}^{n} q_i/n$ and the variance of the sample $\sigma_q^2 = \sum_{i=1}^{n} (q_i - \bar{q})^2/(n - 1)$. The quantity $q$ is said to be $\bar{q} \pm \sigma_q/\sqrt{n}$. This inherently contains the assumption that the sample follows a Gaussian distribution. This is not necessarily the case in low temperature SOP images.

In Figure 5.3, SOP data of shot 32081 and the histograms of three regions of
Figure 5.3: SOP data of shot 32081 and the histograms of three regions of interest. The three regions are: before laser firing, during x-ray exposure, and after compression wave breaking out at the LiF back surface. Before laser firing \((t < 0)\) (green box), every component of the target is at ambient temperature, so no thermal photons are emitted. The received signals are completely due to detector noise, and the histogram is simply a Gaussian distribution. During the x-ray exposure (orange box), the signal levels are low but certainly nonzero, and the histogram seems to be a Gaussian close to the detector noise, but with longer tails. After wave breaks out (red box), many thermal photons are emitted and the histogram is a Gaussian distribution centered at a nonzero value.
interest are shown. Before laser firing \( (t < 0) \) (green box), every component of the target is at ambient temperature, so no thermal photons are emitted. The received signals are completely due to detector noise, and the histogram is simply a Gaussian distribution. After wave breaks out (red box), many thermal photons are emitted and the histogram is a Gaussian distribution centered at a nonzero value. The traditional method applies well to the red region.

During the x-ray exposure (orange box), the signal levels are low but certainly nonzero by human-eye observation, and the histogram appears to be a Gaussian similar to the detector noise, but with tails to the right. It is hypothesized that the signal level is below the detector noise level (Figure 5.1). The majority of pixels do not receive photoelectrons at all, appearing in the histogram as the Gaussian centered at zero. The tail of the histogram corresponds to pixels that are “lucky” to receive some photoelectrons. It is hard to define precisely the mean and uncertainty of \( I \) in this case. Nevertheless, the tail does contain information of the target temperature. In this chapter, we present a framework of inferring target temperature from the non-Gaussian signals.

To give a taste of the model developed in this work (which will be described in Section 5.3), we show in Figure 5.4 some simulated histograms using the model. The histogram is still fairly close to the detector noise Gaussian at 2000 K, but at 3300 K the histogram deviates slightly from the noise. As temperature rises, the histogram approaches a Gaussian centered at nonzero values. A rough inspection implies that the target temperature is close to 3300 K during x-ray exposure for shot 32081, but a more sophisticated and quantitative analysis is required to obtain a precise estimate of \( T \) and its uncertainty.
Figure 5.4: The orange histogram is of the orange region in Figure 5.3. The other histograms are simulated using the model developed in this work at various temperatures. The histogram is still fairly close to the detector noise Gaussian at 2000 K, but at 3300 K the histogram deviates slightly from the noise. As temperature rises, the histogram moves to higher $I$ and approaches a Gaussian centered at nonzero values. A rough inspection implies that the target temperature is close to 3800 K, but a more sophisticated and quantitative analysis is required to obtain a precise estimate of $T$ and its uncertainty.
5.3 SOP Modeling

In order to understand the behavior of SOP at low signal level, a careful study of the streak camera is required. The streak camera measurement process can be broken down into a few steps and each step introduces additional noises (Figure 5.5) [71]. The thermal photons from the target are collected by the photocathode and converted to an electron beam signal via photoelectric effect. The electron beam is accelerated through the streak tube by an extraction electric field and focused by a focus electrode. The photoelectrons are then deflected by a time-varying electric field before finally striking a phosphor screen. At the phosphor screen, the energetic electrons produce a pulse of light that is transported via a fiber-optic image coupler to a CCD image sensor.

A clarification of notions about pixels is due here. Each pixel on the CCD chip is of area $\Delta A_{\text{CCD}} = (B\Delta x)^2$, if the CCD pixels are binned by $B \times B$. If the source temperature is stationary over a period long compared to the dwell time $\Delta t = W_{\text{LSF}}/B\Delta x\eta$, then there is a one-to-one correspondence between $\Delta A_{\text{CCD}}$, an area on the slit, and an area on the light source. The corresponding pixel size on the slit is $\Delta A_{\text{slt}} = (B\Delta xW_S/W_{\text{LSF}})(B\Delta x/M_{\text{EO}})$. The corresponding pixel size on the light source is $\Delta A_{\text{src}} = (B\Delta xW_S/W_{\text{LSF}}M)(B\Delta x/M_{\text{EO}}M)$. In what follows, the word “pixel” can refer to any of these three objects without causing confusion.

Let the photon spectral number density of the light source be $n(\lambda)$ per pixel. This means, the average number of photons in the wavelength range $\lambda \rightarrow \lambda + \Delta \lambda$ received by each photocathode pixel is $n_p(\lambda)\Delta \lambda$. It is related to the quantities defined in Section 3.3 by

$$n_p(\lambda) = \frac{\Delta t\Delta A_{\text{src}}\Delta \Omega}{hc/\lambda}L_S(\lambda; T)T_x(\lambda)T_r(\lambda)$$

where $T_r(\lambda)$ is the transmission spectra of the combined optical relay, excluding any inserted ND or bandpass filters, which are already accounted for by $T_x(\lambda)$.

The number of photons $N_p(\lambda, \Delta \lambda)$ in this wavelength range per pixel is then
a Poisson variable, with mean value \( n_p(\lambda)\Delta \lambda \). At the photocathode, the incident photons are converted to photoelectrons with a wavelength dependent quantum efficiency \( \varepsilon_q(\lambda) \), the probability that an incident photon produces a photoelectron that will pass through the streak tube. The conversion can be modeled by binomial statistics. The number of produced photoelectrons \( N_e(\lambda, \Delta \lambda) \) in the wavelength range \( \lambda \rightarrow \lambda + \Delta \lambda \) is then another Poisson variable, with mean value \( n_p(\lambda)\varepsilon_q(\lambda)\Delta \lambda \).

The rest of the components of the streak camera does not depend on wavelength, so we can, at this step, combine \( N_e(\lambda, \Delta \lambda) \) into the total number of photoelectrons \( N_e \) over all wavelengths:

\[ N_e = \int \left( \frac{dN_e}{d\lambda} \right) d\lambda. \]

It is a well-known fact that the sum of a finite number of Poisson variables is still a Poisson variable with mean value equal to the sum of mean value of these component Poisson variables. Therefore the number of photoelectrons \( N_e \) generated at the photocathode per pixel follows Poisson distribution,

\[ N_e \sim \text{Poisson}\left( \Lambda = \int n_p(\lambda)\varepsilon_q(\lambda) d\lambda \right), \tag{5.4} \]

where \( \varepsilon_q(\lambda) \) is the wavelength-dependent quantum efficiency of the photocathode.

The photoelectrons are accelerated by the extraction electrode and become very energetic. Multiple CCD counts are recorded for each photoelectron that reaches the phosphor screen, and these counts are distributed over a small area of the CCD. The number of CCD electrons \( N^s \) per photoelectron is again a random variable, whose probability density is \( 72 \)

\[ f_s(N^s) = CN^s \exp\left( -\frac{(N^s - \mu_s)^2}{2\sigma_s^2} \right), \tag{5.5} \]

which lumps together the effects of photoelectron acceleration, fluorescence effect at the phosphor screen, and fluorescence photon to CCD electron conversion process.

It is assumed that each photoelectron generates CCD electrons independently \( 72 \).
The total number of CCD electrons per pixel $N^c$ is a compound Poisson variable,

$$N^c = \sum_{k=1}^{N^e} N^s_k,$$

(5.6)

where $k$ indexes the photoelectrons. It is called “compound” because the number of terms in the sum, $N^e$, is a random variable, and the summands, $N^s_k$, are individually random as well.

The photons produced at a pixel of the phosphor screen (and therefore the CCD electrons at the CCD chip) tend to spread over a small region near the pixel, modeled using a point spread function $g$. The read noise $N^r$ of the streak camera is a Gaussian variable with $\mu_r = 0$ and $\sigma_r$, and the final signal $N^f$ is

$$N^f = N^c \ast g + N^r = \sum_{k=1}^{N^e} N^s_k \ast g + N^r,$$

(5.7)

or in component form,

$$N^f_i = \sum_m g_{i-m}N^c_m + N^r_i.$$  \hspace{1cm} (5.8)

It is understood that the index stands for a pair of indices for temporal location and spatial location of the pixel, i.e. $i = (i_t, i_x)$, and are written in a single variable only for brevity. $g$ in the component form is

$$g_i = \frac{1}{2\pi \sigma_{px}^2} \exp\left(-\frac{i^2}{2\sigma_{px}^2}\right) = \frac{1}{2\pi \sigma_{px}^2} \exp\left(-\frac{i_t^2 + i_x^2}{2\sigma_{px}^2}\right).$$  \hspace{1cm} (5.9)

The intensity $I$ of SOP (in unit of ADU) is related to $N^f$ by $I = G'N^f$, where $G'$ is the gain factor converting CCD electrons to ADUs. Each pixel has a separate measurement of $N^f$. If we assume the temperature of photon source is stationary over a period of time, and is uniform spatially, then $N^f$ is quasi-identically distributed according to the same marginal probability density. The only factor preventing it becoming truly identically distributed is the point spread function $g$, which correlates
neighboring pixels. Essentially, over this period of time and across the target, we obtain a 2D measurement of $N^t$ which contains information about the average number of photoelectrons per pixel, $\Lambda$ (or equivalently, temperature $T$). This is a one-parameter model, and can be solved by maximum likelihood method. The parameter $\Lambda$ is uniquely related to $T$, and is a useful construct and intermediate variable.

At high temperatures, $N^t$ approaches a Gaussian variable by the Central Limit Theorem, so $I = G'(N^t) = G'(N^e)G(N^s) = G'\Lambda(T)G$, where $G = \int_0^\infty N^s f_s(N^s) dN^s$ is the gain of CCD electrons from photoelectrons. Comparing the expression for $\Lambda(T)$ Equation (5.4) and the expression for $I$ Equation (3.7), it is obvious that the system response $SR(\lambda)$ can be related to quantities introduced in this section by

$$SR(\lambda) = \frac{T_q(\lambda)\varepsilon_q(\lambda)}{\hbar c/\lambda}.$$  \hspace{1cm} (5.10)

It is $SR(\lambda)$ that is calibrated absolutely [43], but not $T_q(\lambda)$ or $\varepsilon_q(\lambda)$ individually.
5.4 Statistical Inference

In what follows, assume the measured dataset $N^i$ is a $n_i \times n_x$ matrix, each entry describing the value of $I/G'$ of a pixel. For notational simplicity, rearrange this matrix into a vector $\mathbf{N}^i$ of length $n = n_i n_x$, isomorphic to the original data matrix, with the first $n_x$ entries being the first row of the original matrix, etc.

The goal is to interrogate information about $\Lambda$ from $\mathbf{N}^i$. According to Bayes's theorem, the posterior probability density of $T$, having measured $\mathbf{N}^i$, is

$$Pr(\Lambda|\mathbf{N}^i) = \frac{Pr(\mathbf{N}^i|\Lambda) Pr(\Lambda)}{Pr(\mathbf{N}^i)}.$$  (5.11)

To reflect our ignorance towards $\Lambda$, the prior distribution $Pr(\Lambda)$ can be chosen to be flat for $\Lambda > 0$ and zero for $\Lambda < 0$.

It turns out extremely difficult to obtain an explicit formula for $Pr(\mathbf{N}^i|\Lambda)$ describing both the non-Gaussian-ness of the histogram of $\mathbf{N}^i$, and the correlation among pixels. We can however focus on one at a time and develop two models. Neither of the models can capture the real physics, but are tractable enough to be useful.

5.4.1 Ignore non-Gaussian-ness, but keep interpixel correlations

We first describe a model that ignore the non-Gaussian-ness of the histogram, but focus on the interpixel correlations. Intuitively, at low signal levels, photon events are so rare that two events rarely occur next to each other, hence the interpixel correlation is probably important for picking these photon events out.

The average number of photons $\langle N^i \rangle$ per pixel is

$$\langle N^i \rangle = \langle N^e \rangle \langle N^s \rangle = \Lambda(T) G.$$  (5.12)
The covariance matrix of \( N^t \) is

\[
\Sigma_{IK} = \langle (N^t_I - \langle N^t \rangle)(N^t_K - \langle N^t \rangle) \rangle = g_{IK}\sigma_c^2 + \delta_{IK}\sigma_r^2,
\]  

(5.13)

where

\[
g_{IK} = \frac{1}{4\pi\sigma_{px}^2} \exp\left( -\left(\frac{(I - K)\%n_x}{2\sigma_{px}}\right)^2 + \left(\frac{(I - K)\//n_x}{2\sigma_{px}}\right)^2 \right)
\]  

(5.14)

is the point spread function \( g \) adapted to the vectorized dataset. The symbols \( \// \) and \( \% \) represent the quotient and remainder of the integer division of \( I - K \) by \( n_x \). The variance of \( N^c \) is

\[
\sigma_c^2 = \Lambda(T)(\text{Var}(N^s) + G^2),
\]  

(5.15)

where \( \text{Var}(N^s) \) is the variance of \( N^s \). It should be noted that \( \text{Var}(N^s) \) is different from the square of the parameter \( \sigma_s \) in Equation (5.5), but is easily computed using Equation (5.5).

The hypothesis is that \( N^t \) satisfies a multi-variable Gaussian distribution,

\[
\text{Pr}(N^t|T) = \frac{\exp\left( -\frac{1}{2}(N^t_I - \langle N^t \rangle)(\Sigma^{-1})_{IK}(N^t_K - \langle N^t \rangle) \right)}{(2\pi)^{n_in_t/2} \sqrt{\det(\Sigma)}}
\]  

(5.16)

which ignores the non-Gaussian-ness of the histogram due to Poisson statistics of the component variables, but still captures the interpixel correlations via the covariance matrix \( \Sigma \). This likelihood function depends on temperature \( T \) via the average number of photon \( \langle N^t \rangle \) and covariance matrix \( \Sigma \).

This approximation is conceptually cleaner. If the interpixel correlation vanishes, the measured ADU value of each pixel approximately follows a Gaussian distribution, whose mean is \( \mu_t = \Lambda(T)G \), i.e., the product of the number of photoelectrons generated at the photocathode and the average number of CCD electrons generated by each photoelectron, and whose variance is \( \sigma_r^2 + \sigma_c^2 \), i.e., the read noise of the streak camera and the noise generated by the photoelectron to CCD electron process added.
in quadrature. This is consistent with intuition. The existence of the correlation is intuitively important in low temperature regimes. If very few photoelectrons hit, the photoelectron events should look like pepper-sprays spread across several pixels over the Gaussian background. Given two image with exactly the same histogram, the image with large-value pixels clustered together is more likely to represent an actual SOP image than the image with large-value pixels spread across the image and distant from each other.

However, it is difficult to apply this method numerically in practice, especially when the number of pixels is large. The computer is sometimes overwhelmed by the need to compute the inverse matrix of \( \Sigma \), which is \( n \times n \). I have found that my personal computer often has difficulty handling the case of \( n = 2500 \) pixels.

### 5.4.2 Ignore interpixel correlation, but keep the non-Gaussian-ness

Next we describe a model that ignores the interpixel correlations, but describes the non-Gaussian nature of the histogram. The model, by ignoring interpixel correlations, assumes each pixel gives an independent measurement of a random variable, so the likelihood of measuring the dataset \( \mathbf{N}^t \) is equal to the product of likelihoods of measuring \( N^t \) at each pixel.

Using the method of characteristic functions, we derive a way of computing \( \Pr(\mathbf{N}^t|T) \) exactly:

\[
\Pr(\mathbf{N}^t|T) = \frac{1}{(2\pi)^n} \int e^{-i\zeta^* \cdot \mathbf{N}^t} \varphi_t(\zeta) \, d\zeta
\]

where the integral is over the Fourier variable \( \zeta \), a vector of length \( n \). \( \varphi_t(\zeta) \) is the characteristic function of the random variable \( \mathbf{N}^t \), and is essentially the Fourier transform of \( \Pr(\mathbf{N}^t|T) \). Omitting the details of derivation, \( \varphi_t(\zeta) \) can be computed by

\[
\varphi_t(\zeta) = \prod_{i=1}^n \varphi_r(\zeta_i) \prod_{m=1}^n \varphi_c \left( \sum_{i=1}^n g_{i-m} \zeta_i \right),
\]
where

\[ \varphi_r(\zeta) = \exp\left(-\frac{1}{2}\sigma_r^2\zeta^2\right), \quad (5.19) \]

\[ \varphi_c(\zeta) = \exp(\Lambda(\varphi_s(\zeta) - 1)), \quad (5.20) \]

\[ \varphi_s(\zeta) = \frac{\xi(\mu_s/\sqrt{2}\sigma_s + i\zeta\sigma_s/\sqrt{2})}{\xi(\mu_s/\sqrt{2}\sigma_s)}, \quad (5.21) \]

\[ \xi(z) = 1 + \sqrt{\pi}z e^z [1 + \text{erf}(z)]. \quad (5.22) \]

By hypothesis, neighboring pixels are not correlated, so \( \varphi_t(\zeta) \) can be written in the form of a product, \( \prod_{i=1}^{n} \varphi_i(\zeta_i) \), where

\[ \varphi_i(\zeta) = \varphi_r(\zeta) \prod_{m=1}^{n} \varphi_c(g_{-m}\zeta). \quad (5.23) \]

The no-correlation hypothesis greatly simplifies the numerical computation of \( \Pr(N_t|T} \), which becomes

\[ \Pr(N_t|T) = \prod_{i=1}^{n} \left[ \frac{1}{(2\pi)} \int e^{-i\zeta N_i^t} \varphi_i(\zeta) \, d\zeta \right]. \quad (5.24) \]

The integration becomes the product of \( n \) one-dimensional integrals, instead of the original intractable, \( n \)-dimensional integration, where \( n \) is usually a few thousands.

This is the model that we will actually use.

### 5.4.3 The model transits to the traditional (Gaussian) model in the high \( \Lambda \) limit

A conjecture is made that as \( \Lambda \) grows to infinity, \( N_c \) approaches a Gaussian distribution, with mean \( \mu_c = -i\Lambda \varphi'_s(0) = \langle N^s \rangle \Lambda \) and variance \( \sigma_c^2 = -\Lambda \varphi''_s(0) = \Lambda(\langle N^s \rangle^2 + \sigma_s^2) \).

To verify this, introduce a scaled variable \( \bar{N}_c = (N^c - \mu_c)/\sigma_c \), then the characteristic
function of $\tilde{N}^c$ is

$$
\varphi_c(\zeta) = \exp\left(-\frac{i\zeta\mu_c}{\sigma_c}\right)\varphi_c\left(\frac{\zeta}{\sigma_c}\right) \sim \exp\left[-\frac{\zeta^2}{2} - \frac{\zeta^3\varphi''_s(0)}{6\varphi'_s(0)\sqrt{-\Lambda\varphi''_s(0)}} + \frac{1}{\Lambda}\right]
$$  (5.25)

Ignoring the $1/\Lambda$ term, this is exactly the familiar characteristic function of a Gaussian variable, therefore $\tilde{N}^c$ approaches a standard Gaussian variable asymptotically.

The same is true for $N^t$ as its characteristic function expands to

$$
\ln\varphi_t(\zeta) = i\mu_t\zeta - \frac{1}{2}K_{t,ij}\zeta^2 + \Lambda^{-1/2},
$$  (5.26)

where

$$
\mu_{t,i} = \mu_c \sum_{m=1}^{n} g_{i-m}
$$  (5.27)

$$
K_{t,ij} = \sigma_r^2\delta_{ij} + \sigma_c^2 \sum_{m=1}^{n} g_{i-m}g_{j-m}.
$$  (5.28)

are expectation vector and covariance matrix of the random vector $N^t$. Therefore at the limit of $\Lambda \to +\infty$, $N^t$ asymptotically approaches a multivariate Gaussian variable.

The model in Section 5.3.2 assumes all pixel values satisfy the same distribution. In order to obtain the marginalized probability density function of $N^t$ of a single pixel, let all $t$'s be zero except for one, then

$$
\ln\varphi_t(\zeta) = i\mu_t\zeta - \frac{1}{2}(\sigma_r^2 + \sigma_c^2)\zeta^2 + \Lambda^{-1/2}.
$$  (5.29)

One can easily recognize that $\varphi_t$ is the characteristic function of a Gaussian variable whose mean is $\mu_t$ and variance is $\sigma_r^2 + \sigma_c^2$. 
5.5 Streak Camera Calibration

In order to apply the model developed in this chapter, it is necessary to calibrate a few parameters: the detector noise level $\sigma_r$, the parameters $\mu_s$ and $\sigma_s$ in the distribution function $f_s(N^s)$.

A hundred dark images were taken with the SOP camera shutter closed for various purposes: (1) Without complication due to external photons, these images reveal defects of the CCD chip: hot pixels, pixels less responsive the light than others, defects in all or part of a row or column of the chip; (2) Combining the dark images provides a master dark image with reduced noises as a measurement of the pixel-wise bias level of the CCD chip. If a single dark image is used as the bias measurement instead, the noise of the scientific image, after being bias corrected, will be amplified by $\sqrt{2}$. The noise level of the master dark image is $\sigma_{r0} = 6.435$ ADU.

The task here is to measure how many ADUs are generated at the CCD chip by a single photoelectron generated at the photocathode. An image was taken over 1 sec with the shutter slightly open. The duration and the openness of the shutter is adjusted to ensure that the photons are sparsely populated on the image. Here we describe a statistical method to pick out those pixels hit by a photon and estimate the conversion rate of photoelectrons to ADUs, without relying too much on human judgment.

Each pixel on the image is either hit by a photoelectron or not. The probability of a pixel being hit by more than one photoelectron is vanishingly small, so we discard that possibility as a high order error. There is also possibility that a photoelectron hits the border of two pixels, but the model described here can rule them out. For each pixel on the image, compute the posterior ratio of two models: model $M_0$ assumes no photon hits the pixel, while model $M_1$ assumes exactly one photoelectron at the photocathode at the pixel, with an adjustable parameter $S$ describing the photoelectron-to-ADU conversion rate. The posterior ratio between the two models
CHAPTER 5. LOW TEMPERATURE SOP MEASUREMENTS

is

\[ r = \frac{\Pr(M_0|D)}{\Pr(M_1|D)} = \frac{\Pr(D|M_0) \Pr(M_0)}{\Pr(D|M_1) \Pr(M_1)} \quad (5.30) \]

where \( D \) refers to data. If a pixel is hit by a photoelectron, the pixel should have higher signal than the immediate neighbors, while the immediate neighbors have higher signals than the next-immediate neighbors. The ratio \( r \) is a characterization of the belief which model is truer given the data. If \( r < 1 \), we have confidence that \( M_1 \) is true, i.e., the pixel is hit by a photoelectron, and we use \( \hat{S} \) that maximize the likelihood of model \( M_1 \) as the photoelectron-ADU conversion rate for that pixel.

We acknowledge ignorance of whether \( M_0 \) or \( M_1 \) is true by setting the prior ratio \( \Pr(M_0)/\Pr(M_1) \) to 1. When a photoelectron hits a phosphor screen at some pixel, it spreads its energy over its neighborhood, so for each pixel, we consider the 5 × 5 pixels region surrounding the it. \( \Pr(D|M_0) \) is the likelihood of obtaining data \( D \) assuming \( D \) is just Gaussian noise,

\[ \Pr(D|M_0) = \prod_{i,j=-2}^{2} \frac{1}{\sqrt{2\pi \sigma_r}} e^{-\frac{D_{ij}^2}{2\sigma_r^2}} \quad (5.31) \]

\( \Pr(D|M_1) \) describes the average likelihood of obtaining data \( D \) assuming \( M_1 \),

\[ \Pr(D|M_1) = \int_0^\infty \Pr(D|S, M_1) \Pr(S|M_1) \, dS \quad (5.32) \]

If a photoelectron hits a pixel, it generates \( S \) ADU, spreading over its neighborhood described by the Gaussian kernel \( g_{ij} \), therefore

\[ \Pr(D|S, M_1) = \prod_{i,j=-2}^{2} \frac{1}{\sqrt{2\pi \sigma_r}} e^{-\frac{(D_{ij}-S_{ij})^2}{2\sigma_r^2}} \quad (5.33) \]

Assume the prior \( \Pr(S|M_1) \) is approximately uniform over some interval \([0, \Delta_S]\), and that \( \Delta_S \) is much larger than the width of the likelihood \( \Pr(D|S, M_1) \) near its peak, we
Figure 5.6: The histogram of pixels on the calibration image that are more likely to contain a photon event than just noise. It is fit to $f_s(N^s)$ to obtain an estimation of the parameters $\mu_s$ and $\sigma_s$.

could write [73]

$$Pr(D|M_1) = \sqrt{2\pi\delta_S} \Pr(D|\hat{S}, D),$$ (5.34)

where $\hat{S}$ is the maximum likelihood estimation of $S$ using measured data $D$, at which $Pr(D|S, M_1)$ peaks. In the measurement we conducted, $\Delta_S \approx 350$, and $\delta_S \approx 7 \times 10^{-4}$. The Ockham factor $\sqrt{2\pi\delta_S}/\Delta_S \approx 1 \times 10^6$ therefore severely punishes $M_1$ for having an adjustable parameter.

For each pixel, compute the posterior ratio $r$, and apply the filter $r < 1$ so that only those pixels that are more likely to have been hit by a photoelectron survive. Rank the surviving pixels from low to high. The pixel with the lowest $r$ is the most likely to have been hit by a photoelectron. Starting from lowest $r$, remove possible overlaps: If two pixels close to each other (their $5 \times 5$ neighborhoods overlap) are both hit by photoelectrons, equation Equation (5.33) does not apply. For such a pair, replace the pixel with higher $r$ along with its $5 \times 5$ neighborhoods with NaN’s. For the surviving pixels, collect their $\hat{S}$ and fit the histogram to Equation (5.5).
The fit result and covariance matrix for the vector $(\mu_s, \sigma_s)$ are

$$
\begin{pmatrix}
84.92 \\
38.75
\end{pmatrix},
\begin{pmatrix}
37.4671 & -13.0423 \\
-13.0423 & 4.68561
\end{pmatrix}.
$$

(5.35)

respectively. The gain of the streak camera, $G$, is

$$
G = \frac{\int N_s f_s(N_s^2) \, dN_s}{\int f_s(N_s^2) \, dN_s} = 102.4 \pm 3.2.
$$

(5.36)

This is consistent with the factory value for $G$, which is $\sim 100$.

5.6 Application to Simulated Data

By following the steps outlined in Section 5.3, one can create simulated samples of $N^I$ by specifying a true $\Lambda_{\text{true}}$. Once the sample is created, one forgets about $\Lambda_{\text{true}}$ and uses the model developed in Section 5.4.2 and the traditional statistics method (described at the beginning of Section 5.2) to infer the parameter $\Lambda$ and its uncertainty from the simulated sample. The result is shown in Figure 5.7 for $\Lambda_{\text{true}} = 0.01$ (corresponding to $T_{\text{true}} = 2936$ K for the new OMEGA EP SOP) over a region of $20 \times 125$ pixels. It is evident that the posterior distribution of $\Lambda$ using the new model is very close to a Gaussian distribution, and the results of the two methods are almost identical. This shows that at $\Lambda_{\text{true}} = 0.01$, the traditional method works nicely and is sufficient. It is also evident that the confidence interval does not necessarily cover the true $\Lambda_{\text{true}}$.

If better temporal or spatial resolution is desired, one might use fewer pixels for the inference. Figure 5.8 shows six samples with $\Lambda_{\text{true}} = 0.01$ over a region of $1 \times 125$ pixels. It is evident that lowering the number of pixels gives rise to the asymmetry of the posterior distribution of $\Lambda$ in the new model. The new model correctly captures the asymmetry and the inferred confidence interval does not cover negative $\Lambda$ values, which is to be expected based on physical considerations. In contrast, the traditional
Figure 5.7: Application of the new and traditional models to the simulated samples with $\Lambda_{true} = 0.01$ over $20 \times 125$ pixels. The blue curve is the posterior density function of $\Lambda$ of the new model. The blue vertical line indicates the maximum likelihood estimation of $\Lambda$ in the new model, while the blue band indicates the shortest interval that covers 68% probability. The orange line and band represents those using the traditional statistics model. Six samples are examined.
Figure 5.8: Application of the new and traditional models to the simulated samples with $\Lambda_{\text{true}} = 0.01$ over $1 \times 125$ pixels.
model covers negative $\Lambda$ values, as is evident in Figure 5.8(b)(c). Therefore in the case of $\Lambda_{\text{true}} = 0.01$ over $1 \times 125$ pixels, the new models still work well and is more robust than the traditional method.

As a final and more extreme example, let us examine the case of $\Lambda_{\text{true}} = 0$ over a region of $20 \times 125$ pixels, that is, apply both models to samples with absolutely no signals at all. The result is shown in Figure 5.9. This shows further the advantage of the new model over the traditional model. In Figure 5.9(a), the sample average is negative, so the traditional method gives a confidence interval that is entirely negative, which is clearly absurd. In Figure 5.9(a)(b)(e), the new model shows that $\Lambda$ is most likely to be 0, but has a some chance to take a positive value, capped above by the model. However, one should beware that it is completely possible that the background noise of some pixels are so “luckily” high that they are mistaken as signals even by the new model. Figure 5.9(c) shows an example of this phenomenon, where the confidence interval given by the new model does not even cover 0. This shows that care should always be taken when one works with statistics.

In summary, we presented a statistical model for interrogating SOP images for low-temperature experiments. Low temperature in this context means temperatures below 4000 K. The components of the SOP, including the streak camera, are analyzed, together with several noise generating mechanisms. A proposed numerical method for maximum likelihood estimation gives robust confidence intervals for simulated low signal-to-noise ratio samples compared to traditional techniques. This sets the stage for analyzing future low temperature pyrometry data.
Figure 5.9: Application of the new and traditional models to the simulated samples with \( \Lambda_{\text{true}} = 0 \) over 20 \( \times \) 125 pixels, that is, samples that are purely background noises with no signal at all.
Chapter 6

Conclusion

At extreme pressures, the interatomic distance in many substances is so small that the inner electrons of atoms constituting the material contribute to the peculiar and often unexpected physical and chemical properties of the material. The development of computational methods have given rise to predictions of phase diagrams of numerous materials at extreme conditions, whose validity is to be benchmarked by experiments, while simultaneously providing a guidance for experiment design. Recently, development of dynamic compression techniques and related diagnostics enables the exploration of extreme conditions that were previously unreachable. By measuring the thermodynamic variables of a material at various extreme states, one gains a benchmark against theoretical predictions. The demand for better measurement of these extreme states also encourages development of new diagnostics and improvement of existing ones.

Silicon is one of the most abundant elements on Earth, found in sands, rocks, semiconductor chips. It is also an important constituent of cores of Earth and earth-like planets. By measuring the phase diagram of silicon at extreme conditions, the behavior of these planets can be better modeled. Silicon has been studied extensively under compression at ambient temperature, exhibiting a rich collection of phase transitions up to 248 GPa. Shock experiments have explored a variety of phase transitions in Si to 54 GPa. Recent first-principle simulations predict that a new dhcp phase between 22 and 55 GPa along the principal isentrope, and remains stable as
fcc phase to 2.8 TPa.

The experiment described in this thesis explores the phase diagram of silicon near its isentrope from 40 to 400 GPa, by ramp compressing silicon by a laser drive. Thermodynamic states of silicon at these states are measured by VISAR, PXRDIP, and the crystal structure is determined by PXRDIP. The experiment shows a significant increase of the stability range of the Si hcp phase compared to theoretical predictions. The hcp phase is observed at the pressure and temperature range where dhcp phase was predicted, and no evidence of the dhcp phase is observed. Furthermore, the hcp-fcc phase transition pressure is at least 93 GPa, much higher than the 55 GPa predicted by computation. This observation is consistent with previous shock compression experiments [60]. The fcc phase is confirmed to remain stable to at least 400 GPa.

Currently, no temperature data exist from nanosecond pyrometric measurements on ramp compression experiments. Such measurements are difficult due to the low number of photons emitted from low temperature (lower than 4000 K) targets. In this work, we present the foundational framework for analyzing low signal-to-noise ratio data. This method yields identical results as traditional techniques at high temperatures, but is more robust at low temperatures. This sets the stage for analyzing future low temperature pyrometry data.
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Chapter A

Si X-ray Diffraction Data Tables

A.1 Experimental Parameters and Results

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<th>XRS</th>
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<th>Δ (ns)</th>
<th>β (µm)</th>
<th>P (GPa)</th>
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Table A.1: Experimental parameters and results of x-ray diffraction experiments of ramp compressed silicon. The columns are described in the main text.

This table contains vital information of the ten shots discussed in Section 4.3. The columns are:

1. Shot number.
2. Structure of the silicon sample at the time of x-ray probing.
3. Pinhole substrate material.
4. Pinhole substrate diameter (µm).
5. X-ray source material.
6. X-ray wavelength (Å).
7. Target components and their thicknesses. For example, C[20][Si][20][LiF][100] means the target is composed of 20 µm thick diamond ablator, 20 µm thick silicon sample, and 100 µm thick lithium fluoride window. The laser hits the target from the “left side”, i.e., on the diamond ablator. The pieces are held together by 1 µm to 2 µm glue layers.
8. Time of 1-ns x-ray pulse relative to the start of the ramp drive (ns).
(9) Sample thickness during the x-ray exposure (μm), inferred from hydrodynamic
simulations and corroborated by VISAR measurements. The sample thickness
is critical to correctly project the image plate pixels to the $2\theta$-$\phi$ space viewed
from the sample. It is defined as the distance from the center of the sample to
the center of the pinhole at the time of x-ray exposure, i.e., the sum of (i) half
of thickness of compressed sample, (ii) thickness of compressed LiF window,
and (iii) half of uncompressed pinhole substrate. The notation $z_s$ is consistent
with section C of [34].

(10) Average sample pressure and its uncertainty during the x-ray exposure (GPa).

(11) The 1-$\sigma$ interval of pressure distribution characterizing the pressure non-
uniformity during the x-ray exposure (GPa).

(12) The lattice parameter $a$ of either hcp or fcc structure (Å).

(13) The axial ratio $c/a$ for hcp structure.

(14) The sample density and its uncertainty (g/cm$^3$).

### A.2 X-ray Diffraction Data

These tables contain x-ray diffraction peak locations $2\theta_{\text{exp}}$ and interplanar distances $d$ of all experiments. They are compared with $2\theta_{\text{fit}}$ calculated using the best-fit lattice parameters in Appendix A. Some of the sample lines, while present in the diffraction image, overlap with pinhole lines, therefore were not used in the fit.

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<th>(100)</th>
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<th>(004)</th>
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<td>78.243</td>
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Table A.2: X-ray diffraction data for shots where sample is identified as hcp structure.
*For shot 32081, there are faint signals at around 86.2° with width 1.1° and 90.7°
with width 1.8°. They are in rough agreement with the predicted locations of (200),
(112), (004) and (201) peaks using the best-fit lattice parameters. These lines are not
well resolved and are not used for the fit.
<table>
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<th>Shot</th>
<th>Miller Index (fcc)</th>
<th>(111)</th>
<th>(200)</th>
<th>(220)</th>
<th>(311)</th>
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Table A.3: X-ray diffraction data for shots where sample is identified as fcc structure. ** Overlap with Pt pinhole line fcc-(002), therefore not used for the lattice parameter fit. The possible overlap explains why the interpreted fcc structure of the sample does not produce more lines. † Overlap with W pinhole line bcc-(002). ‡ Overlap with W pinhole line bcc-(002) and bcc-(222) respectively.