Contents

[**Chapter 1:** Introduction 2](#_Toc72848790)

[1.1 The Dust cycle in the Galaxy 2](#_Toc72848791)

[1.2 Ice formation in molecular clouds 2](#_Toc72848792)

[1.2.1 Observations 2](#_Toc72848793)

[1.2.2 Vapor deposited Amorphous ice experiments 2](#_Toc72848794)

[1.2.3 Ice modelling 2](#_Toc72848795)

[1.2.4 Synergy between Experiments, Modelling and Observations in Astronomy 2](#_Toc72848796)

[1.3 Ice processing during star and planet formation 3](#_Toc72848797)

[1.3.1 Star Formation, Step by step 3](#_Toc72848798)

[1.3.2 Protoplanetary Disks 3](#_Toc72848799)

[1.3.3 Planet formation processes 3](#_Toc72848800)

[1.3.4 Collision Experiments 3](#_Toc72848801)

[1.4 Ices in the Solar system 3](#_Toc72848802)

[1.5 Ices in the laboratory 4](#_Toc72848803)

[1.5.1 Water phase diagram 4](#_Toc72848804)

[1.5.2 Crystalline vs Amorphous ice 4](#_Toc72848805)

[1.5.3 Water polyamorphism 4](#_Toc72848806)

[1.6 The Science of phase transition 4](#_Toc72848807)

[1.6.1 Liquid to HGW 4](#_Toc72848808)

[1.6.2 ASW to crystalline ice 4](#_Toc72848809)

[1.7 Thesis motivation and aim 5](#_Toc72848810)

[1.8 Thesis Outline 5](#_Toc72848811)

[**Chapter 2:** Theory and Methods 5](#_Toc72848812)

[2.1 IR Spectroscopy 5](#_Toc72848813)

[2.2 Neutron Scattering 5](#_Toc72848814)

[2.2.1 Scattering Theory 5](#_Toc72848815)

[2.2.2 NIMROD Instrument 5](#_Toc72848816)

[2.2.3 Limitations 5](#_Toc72848817)

[2.3 Acoustic Levitation 6](#_Toc72848818)

[2.4 The Portable Astrochemistry Chamber 6](#_Toc72848819)

[2.4.1 Setup description 6](#_Toc72848820)

[2.4.2 Experimental procedure 6](#_Toc72848821)

[2.4.3 Data acquisition 6](#_Toc72848822)

[2.4.4 Data Reduction 6](#_Toc72848823)

[2.4.5 Data Processing 6](#_Toc72848824)

[**Chapter 3:** Investigation of bulk amorphous water ice structure, morphology, and crystallisation kinetics 7](#_Toc72848825)

[3.1 Introduction 7](#_Toc72848826)

[3.1.1 Literature study 7](#_Toc72848827)

[3.1.2 Challenges resulting from the metastable structure of ASW 7](#_Toc72848828)

[3.1.3 Sample overview 7](#_Toc72848829)

[3.2 Deposition scans 7](#_Toc72848830)

[3.2.1 20 Kelvin depositions 7](#_Toc72848831)

[3.2.1 Depositions at higher temperatures 7](#_Toc72848832)

[3.3 Annealing 7](#_Toc72848833)

[**Chapter 4:** Influence of impurity (ethane) on the structure, morphology, and crystallisation kinetics of ASW 7](#_Toc72848834)

[**Chapter 5:** Formation and investigation of the optical properties and aggregation of levitated icy micro-particles 8](#_Toc72848835)

[**Chapter6:** Novel methodology for producing ice micro-particles 8](#_Toc72848836)

[**Chapter 7:** First characterisation results of ice micro-particles 8](#_Toc72848837)

[**Chapter 8:** Summary and future directions 8](#_Toc72848838)

## **Chapter 1:** Introduction

## The Dust cycle in the Galaxy

Dust grain produced in supernovae (Carbonaceous/Silicate) – Surface for molecule to condense

Molecules destroyed by Interstellar radiation field

## Ice formation in molecular clouds

Grains shielded from interstellar Radiations, Atoms condense and react to form ice layer

### 1.2.1 Observations

**Infrared (Vibration):** Spitzer/ Hershel/ Sofia … JWST

**Radio (Rotation):** ALMA

Molecular diversity 🡪 Astrochemistry (200 molecules detected)

Highlight the need

### 1.2.2 Vapor deposited Amorphous ice experiments

Amorphous Solid Water (ASW) – Vapor Deposition - Porosity, Gas trapping …

Lab Experiments / Ice Modelling with respect to Ice observations described before !

* **What we know – what remain to investigate**

### 1.2.3 Ice modelling

How to investigate ice formation. Various description of the water molecules …

### 1.2.4 Synergy between Experiments, Modelling and Observations in Astronomy

Dust optical properties n and k value 🡪 Synthetic spectra 🡪 Fit observations

Limitations: n and k values for 100% Amorphic or 100% Crystalline. How is the mixture modelled ?

## Ice processing during star and planet formation

### 1.3.1 Star Formation, Step by step

Protostar - Class 1 – Class 2 – Class 3 – Debris

Explain each phase using Spectral Energy Distribution (SED) – highlight the many parameters used in fittings SED with dust properties.

Is ice reprocessed ? How much

### 1.3.2 Protoplanetary Disks

Birthplace of planets

Structure – Snowline

ALMA Observations - substructure

Turbulence

### 1.3.3 Planet formation processes

Different theory …

### 1.3.4 Collision Experiments

Planet formation – collision experiments – Bouncing barrier – no ASW aggregate produced to perform collisions

Role of ice phase in planet formation

## Ices in the Solar system

Interplanetary dust – Remnants of primordial ice ?

Comets (D/H ratio) – Ice + impurities – track history of planet formation

Icy Moons – Enceladus, Europa – High density amorphous ices

## Ices in the laboratory

### 1.5.1 Water phase diagram

Investigation techniques (spectroscopy, scattering, DSC …)

What do we know 🡪 What are the gaps?

### 1.5.2 Crystalline vs Amorphous ice

20 crystalline forms + 5 amorphous + supercooled liquid etc …

### 1.5.3 Water polyamorphism

Start by showing history of misconceptions

Build up my own classifications

1. Low density
   1. ASW
   2. HGW
   3. LDA
2. High Density
   1. HDA
   2. vHDA

Difference in properties between each of them – Why does this matter ?

## The Science of phase transition

### 1.6.1 Liquid to HGW

High cooling rate to avoid critical nucleus (crystallites) formation

Water = poor glass former 🡪 complicated to achieve, hard to investigate experimentally

### 1.6.2 ASW to crystalline ice

Use isothermal to probe crystallisation kinetics and make link with above

Easier to achieve experimentally

Describe both state with cooperative Hydrogen Bonding networks

Glass transition

## 1.7 Thesis motivation and aim

## 1.8 Thesis Outline

## **Chapter 2:** Theory and Methods

## 2.1 IR Spectroscopy

Good for molecular composition but limited for structural information

## 2.2 Neutron Scattering

### 2.2.1 Scattering Theory

Structural Information

Why Neutrons and not X-Ray

Small Angle – Large angle – Wide angle

### 2.2.2 NIMROD Instrument

Wide angle – probe both surface and bulk properties

### 2.2.3 Limitations

Lots of material required

Heavy sample transfer handling – not suitable for metastable ice

Container – source of heterogeneous nucleation

## 2.3 Acoustic Levitation

Standing wave acoustic levitation

## 2.4 The Portable Astrochemistry Chamber

### 2.4.1 Setup description

1. Describe all components (Surface, pumps, IR spectrometer, Optics)

### 2.4.2 Experimental procedure

### 2.4.3 Data acquisition

1. Ice growth and thickness measurement
2. Omnic software
3. Annealing
4. Data recovery and storage

### 2.4.4 Data Reduction

1. Reduction Strategy
2. DR1: Sanity Check / Merging
3. DR2: Baseline Correction
4. DR3: Integration

### 2.4.5 Data Processing

1. Processing Strategy
2. DP1: Subtraction Scans
3. DP2: Gaussian fitting

## **Chapter 3:** Investigation of bulk amorphous water ice structure, morphology, and crystallisation kinetics

## 3.1 Introduction

### 3.1.1 Literature study

### 3.1.2 Challenges resulting from the metastable structure of ASW

### 3.1.3 Sample overview

Table of all samples

Justifications for each of them

Find a proper Classification and explain

## 3.2 Deposition scans

### 3.2.1 20 Kelvin depositions

### 3.2.1 Depositions at higher temperatures

### 

## 3.3 Annealing

## **Chapter 4:** Influence of impurity (ethane) on the structure, morphology, and crystallisation kinetics of ASW

## **Chapter 5:** Formation and investigation of the optical properties and aggregation of levitated icy micro-particles

## **Chapter6:** Novel methodology for producing ice micro-particles

## **Chapter 7:** First characterisation results of ice micro-particles

## **Chapter 8:** Summary and future directions