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School of Engineering and Sciences

Master of Science in Nanotechnology
Thesis Proposal

Computational Study of Exotic and Glassy Behaviour in Decimated Square Colloidal Ice

by

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The committee members, hereby, recommend that the proposal presented by Leonardo Gabriel Alanis Cantu to be accepted to develop the thesis project as a partial requirement for the degree of **Master of Science in Nanotechnology**.

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1 Introduction

Glassmaking has a rich history dating back over 4000 years, first being discovered near the River Belus in Syria, and then produced by craftmen using simple materials such as silica, alkali and fire [1]. Over millennia, glassmaking techniques have evolved dramatically, resulting in a wide array of glass types, from ancient stained glass to modern optical fibers and smartphone screens. Today, the structure of various types of glass is well characterized, thanks to advancements in materials science and analytical tools. However, despite this progress, the mechanism responsible for a substance transitioning into a "glassy" state during the so-called *glass transition* is not well understood [2].

Understanding the glass transition is critically relevant in various scientific, industrial, and everyday contexts. At its core, the glass transition is a change in behaviour that materials undergo when cooled from a liquid state to a solid state without crystallizing. This transition has significant implications for material properties and behaviors, potentially allowing for the design of novel materials with tailored properties and enhanced performance.

Given that glasses are neither solids nor liquids, but share properties of both, they are seldom described in detail by condensed matter and solid state physics books. Modern theoretical approaches to the glass transition date to the 70s with Anderson's work on the Theory of Spin Glasses [3], first solved analytically in the 80s. Later theoretical developments such as mode-couplig theory and kinetic glass lattice models date to the mid to late 80s[4].

In broad terms, a material is *glassy* when its relaxation time becomes much larger than the duration of an experiment or a simulation. Having a broad definition lets us classify many materials as glasses e. g. window panes, hard systems such as superconductors, or within the scope of this work, soft condensed matter systems such as colloidal systems. Rich phenomenology has been found in glassy materials such as aging, hysteresis, creep memory, non-linear response etc. hence an increased interest in researching a broad range of glassy materials.

Recent studies have shown that glassy behaviour can appear in colloidal systems at the single particle level [5] and a strong correlation between static structure and soft modes with the slowing down of dynamics [6] in buckled colloidal monolayers. Additionally, defects in lattices are suggested to play an important role in the slowing down of dynamics as the glass transition is approached [7], emphasizing the need to consider structural imperfections when exploring glassiness.

Colloidal ice represents a system that emulates artificial spin ice. In this setup, paramagnetic colloidal particles find themselves restricted within double well traps, brought about by gravity and influenced by an external magnetic field that imparts magnetic moments to these particles. This concept, originally introduced by Libal et al. [8], was later experimentally actualized by Ortiz-Ambriz and Tierno [9]. Remarkably, this system recovers yields the ice rule as an emergent phenomenon, driven by strong dipole-dipole interactions.

The primary objective of this research is to investigate the possibility of observing glassy behaviour in square colloidal ice through decimation using Molecular Dynamics simulations. In doing so, the research aims to contribute to a deeper understanding of the underlying principles governing the glass transition in complex systems. One of expected achievements is the identification of evidence pointing towards the existence of slow dynamics within the system, and perhaps novel behaviour.

The document is organized as follows. In 1.1 the problem is defined in detail, section 1.2 states the hypothesis and the research questions to be answered. In section 1.3 the general and particular objectives of this research, as well as the scope of this work are defined. Section 2 contains the relevant background, and theoretical framework. Section 3 elaborates on the methodology of this research, and 4 contains general aspects to the workplan to be followed.

1.1 Problem Statement and Context

The glass transition, a phenomenon observed in materials like glass, polymers, and some liquids when a transitions from a supercooled liquid to an amorphous solid state as it is cooled below a certain temperature T_g below its melting point characterized by a slowing down of dynamics, and an increase in the structural relaxation time τ_α. While the temperature for this transition is well-documented for certain polymers [10], there are still numerous unanswered questions surrounding it.

First, as a continuous transition, the glass transition temperature T_g is ill-defined, it depends on the cooling rate, meaning that it is not a fundamental property, but a property depending on the thermal history of the material. Secondly, the *transition* as a characteristic, is poorly understood. Although it seems to be a second order phase transition, some derivatives are not as sharp as in a true second order transition. Moreover, glass does not necessarily correspond to symmetry change, but to an example of broken ergodicity. The glass structure is whatever the liquid finds itself in. [2]. The main question on the glass transition is whether it corresponds to a thermodynamic or a dynamical phenomenon [11].

A glass transition is typically described in terms of relaxation times τ_α greater than the typical observation time τ_0. For colloids, governed by Brownian motion, this timescale is set by the time to diffuse a diameter

$$\tau_0 = \tau_B = \frac{3\pi\eta\sigma^3}{k_B T}, \tag{1.1}$$

where η is the solvent viscosity. Relatation times can be fitted experimentally from a temporal correlator $C(t) = \langle \hat{C} \rangle$ where $\hat{C}$ is the spatial average of a mobility field $c(\mathbf{r}, t)$. This correlator typically follows the Kohlrausch-Williams-Watts law

$$C(t) = C \exp\left[-\left(\frac{t}{\tau_\alpha} \right)^b \right], \tag{1.2}$$

with $0 \leq C \leq 1$, and $0 \leq b \leq 1$. Further measurements include spatial heterogeneity of the dynamics through spatial correlation functions of the mobility field.

$$G(\mathbf{r}, t) = \langle \delta c(0,0) \delta c(\mathbf{r}, 0) \rangle \,. \tag{1.3}$$

Similar to critical phenomena, the spatial correlation should decay as an exponential as the glass transition is approached

$$G(r, t) \sim \frac{1}{r^p} e^{-r/\xi(t)}, \tag{1.4}$$

where ξ is the correlation length. The main difference with typical criticality is that in glass, one deals with a mobility field, instead of a static field [11].

Previous studies show that glassy behaviour can appear in colloidal systems [5] and conclude that parallels can be made between a static structure and soft modes with slow dynamics [6] in buckled colloidal monolayers. Additionally, defects in lattices are suggested to play an important role in the slowing down of dynamics as the glass transition is approached [7]. Understanding the glass transition has broad and significant implications across various fields, leading to numerous potential applications. In materials science and engineering, a comprehensive grasp of the glass transition can aid in the development of advanced materials with tailored properties, including high-performance polymers, glasses, and ceramics.

1.2 Hypothesis and Research Questions

The introduction of glassy behavior is achievable within square colloidal ice systems through the process of decimation. Decimation, characterized by the deliberate introduction of disorder compared to the ground state, impedes the spontaneous crystallization process. This introduced disorder has the potential to foster

heterogeneity within the system, facilitating the emergence of cooperative domains and regions exhibiting glassy behavior

The research questions that are going to be addressed are:

- How does the degree of decimation affect the emergence of glassy dynamics in square colloidal ice?
- Can we identify a correlation between the spatial distribution of missing colloids and the formation of glassy states in the system?
- Can we experimentally observe slow relaxation processes, characteristic of glassy materials, in decimated colloidal ice samples?
- Can we draw parallels between the behavior of decimated square colloidal ice and other glass-forming systems, such as supercooled liquids or amorphous solids?
- How does the presence of topological defects (e.g., dislocations) in the colloidal ice lattice influence glassy dynamics in decimated systems?

1.3 Objectives and Scope

The general objective of this research is to investigate using overdamped Brownian dynamics simulations the conditions under which glassy and novel behaviour can be induced in square colloidal ice through the deliberate decimation.

The particular goals to achieve, as this research is conducted are the following:

- Simulate colloidal ice with anisotropic forces by changing the alignment of the external magnetic field with respect to the z axis.
- Program a simulated annealing algorithm to find a ground state of unknown systems.
- Simulate decimated colloidal ice with a controlled fraction of decimation f.
- If exists, determine the minimum fraction of removed colloids f_g, and external magnetic field B_g required to induce glassy behavior.
- Compute and analyze spatial correlation functions to examine heterogeneity in the decimated lattice, as well as to identify possible cooperative regions.
- Calculate and compare relaxation times in decimated and non-decimated systems to assess the effect of disorder on dynamics.
- Investigate the formation and evolution of cooperative domains within the decimated lattice.

2 Theoretical framework

2.1 Geometrical frustration

Geometrical frustration is a phenomenon that arises in many physical systems when local interactions cannot be satisfied simultaneously. Consider as an example the Ising model in a square lattice with the Hamiltonian

$$H = -\sum_{\langle i,j \rangle} J_{ij} s_i s_j, \tag{2.1}$$

with antiferromagnetic (AF) interactions $J_{ij} < 0$, $\sigma_i = \pm 1$, and $\langle i, j \rangle$ denotes sum over nearest neighbours. It can be shown that the ground state corresponds to anti parallel neighbouring spins. However, in a triangular lattice when an AF bond is satisfied, the third spin cannot be AF bonded with both and has at least one frustrated interaction. It has been shown that the ground state of the AF Ising model in a triangular lattice correspond to each spin having two frustrated bonds [12] (see figure 1) In essence, the richness of frustrated systems comes from achieving non-trivial configurations due to its topology and nature of the interactions, highly degenerate ground states and residual entropy at low temperatures.

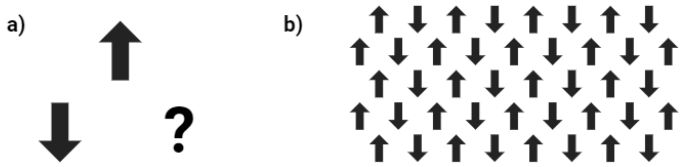

Figure 1: Ising spins in a triangular lattice. (a) Three spins with AF bonds cannot satisfy all interactions simultaneously. (b) Ground state of the Ising model in a triangular lattice.

Frustrated systems have been extensively researched both theoretically, and experimentally in many disciplines. In the last decade, due to advancements in lithographic techniques, the engineering of frustrated systems has taken relevance, particularly in magnetic materials and artificial spin ice [13] that lead to novel emergent properties such as magnetic monopoles [14, 15], Coulomb phases [16, 17], and glassy behaviour [5–7].

2.2 The Ice Rule

Water has some fascinating properties that are almost unique to it, e. g. water reaches its maximum volume at 4 °C. One particular property of water at low temperature is its residual entropy, implying a degenerate ground state at zero Kelvin. This characteristic was first evidenced by Giauque and Ashley in 1933 [18] where they performed careful calorimetric experiments to deduce the entropy of ice and found that it was not zero in spite of its crystal structure. Later in 1935 Pauling [19] confirmed this behaviour when he calculated the residual entropy of water ice both theoretically and experimentally. An elegant and simple explanation came from the ice rule of Bernal and Fowler from two years before [20]. Simply, at low temperatures each water molecule is oriented such that the hydrogen atoms point towards two of the four neighbors, forming hydrogen bonds (see figure 2). Pauling showed that the number of configurations for this arrangement grows exponentially with the number of oxygen atoms. And thus results in an extensive degeneracy which produces residual entropy.

The arrangements of hydrogen bonds in water ice can be described in terms of magnetic dipoles, i. e. the arrangement of ice, also known as the pyrochlore lattice, can be replaced entirely by a spin system where the spins lie on the edges of the lattice and point either inwards or outwards of the hydrogen atom *(vertex) depending on whether the proton is close or far (figure 2) [21]. This this 2in/2out spin arrangement of magnetic moments in the pyrochlore lattice is what is often referred as the *Ice Rule*. However, this characteristic is generalized in section 2.2.1.

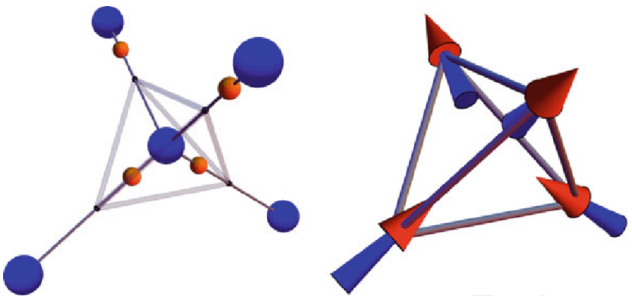

Figure 2: Ice rule for water ice. Water molecules (left) form hydrogen bonds, each hydrogen atom is oriented in such a way that bonds with the two nearest oxygen atoms. Atoms can be replaced by spins (left) pointing in or out depending on whether protons are close or far. On this arrangement, there are two spins in, and two spins out. Image by Nisoli [21]

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2.2.1 Spin Ice, Artificial Spin Ice, and Topological Concepts of the Ice Rule

At low temperatures, the behavior of the spins in spin ice materials becomes highly structured and can be effectively modeled as binary variables, akin to classical Ising spins. These spins align themselves along the bonds of the pyrochlore lattice, adopting either *up* or *down* orientations. This simplification allows the complex three-dimensional spin ice lattice to be projected into two dimensions, effectively creating a square lattice. On this simplified lattice, a specific set of possible spin configurations emerges, where each spin can interact with its neighboring spins in accordance with well-defined rules.

Within this reduced framework, there are six distinct types of local vertices, and these are degenerate in terms of their rotational symmetry. These vertices represent the various ways in which spins can be arranged at their meeting points on the square lattice. This model has been shown to recover the fundamental properties of water ice such as extensive degeneracy, frustrated interactions, and most importantly the ice rule [22].

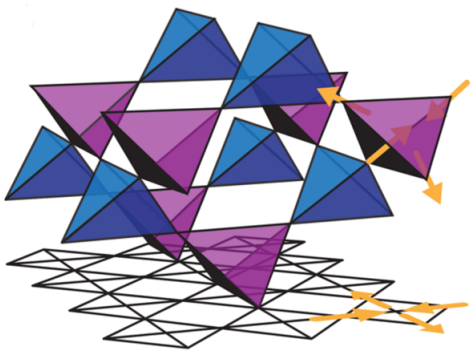

Figure 3: Spin ice in a square lattice. The pyrochlore lattice can be projected into a square lattice. Image by Glaetzle et. al. [23]

The ice rule can be explained in more general terms. Consider a general lattice with Ising spins pointing along the edges. At any given vertex with coordination number z and n spins pointing towards it, then we can define a topological charge q as

$$q = 2n - z, \tag{2.2}$$

which corresponds to the difference of spins in-out. Considering the ice rule (2in/2out), then it is easy to see that this ground state has no topological charge. Note that a flip of any given spin will alter the topological

charges of its neighbours. Using this notion, any configuration that locally minimizes $|q|$ obeys the ice rule, and a subset of phase space of such configurations is regarded as the ice manifold.

Note that in a uniform lattice, such as the square lattice, the $q = 0$ ice rule has two possible configurations: two opposite spins inwards, and two neighbouring spins inwards. Violations to the ice rule come in the form of topological monopoles with $q = \pm 2$ (3in/out, and 1out/in), and $q = \pm 4$ (4in and 4out). However, not all lattices allow local zero charge, e. g. in odd coordination lattices such as the Kagomé lattice cannot be done, and in mixed coordination numbers, several versions of the ice rule can appear [22].

2.2.2 Artificial Spin Ice

Artificial spin ice is a man-made metamaterial, a nanoscale version of the spin ice materials found in nature, designed and engineered for scientific research with potential applications in data storage and magnetronics. In artificial spin ice, magnetic nanostructures, typically nanomagnets, are precisely patterned into arrays of geometric shapes that can form square, triangular, Kagomé (see figure 4), Cairo and other more exotic lattices. Unlike natural spin ice, artificial spin ice allows for precise control and manipulation of individual spins, making it a versatile platform for studying magnetic phenomena and potential applications.

One of the key advantages of artificial spin ice is its tunability. Researchers can design and engineer the geometry and interactions between the artificial spins, enabling them to create tailored magnetic landscapes with specific properties. This control has led to the observation of a wide range of magnetic behaviors, including emergent phenomena like magnetic monopole-like excitations, chiral dynamics and phase transitions [24].

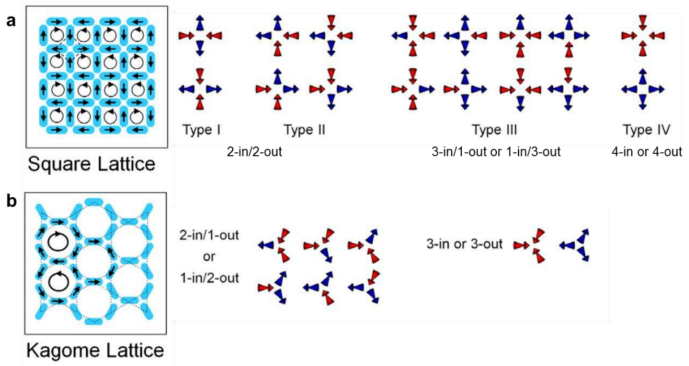

Figure 4: Typical arrangements and vertices for Artifical Spin Ice systems. (a) In a square lattice there are four types of vertices where type I and II correspond to the Ice Rule. (b) In a Kagomé lattice there are two types of vertices only. Image by Skjærvø et. al. [24]

2.3 Colloidal Ice

Colloidal ice refers to a unique and intriguing experimental system that mimics the behavior of atomic and molecular water ice but is composed of microscopic colloidal particles suspended in a fluid. What makes colloidal ice particularly fascinating is that it allows researchers to directly observe and manipulate the dynamics of individual particles in real time.

Influenced by the advancements in Artificial Spin Ice, it was proposed that a lattice made by double well traps where each trap can hold up to one colloidal particle, could mimic the behaviour of spin ice systems. A spin degree of freedom can be mapped on this system pointing towards the well in which the colloid is trapped (see figure 5) [22]. Historically, simulations of this system came first by Libál et. al. [8] where

Brownian Dynamics simulations where used, and colloids consisted of charged particles with a screened Coulomb potential for colloid-colloid interactions. Although the local ground state does not correspond to the ice rule (4out for colloids), frustrated colloid-colloid interactions as the system is strongly coupled recover the ice rule (2in/2out) at the single vertex level as an emergent property of the system.

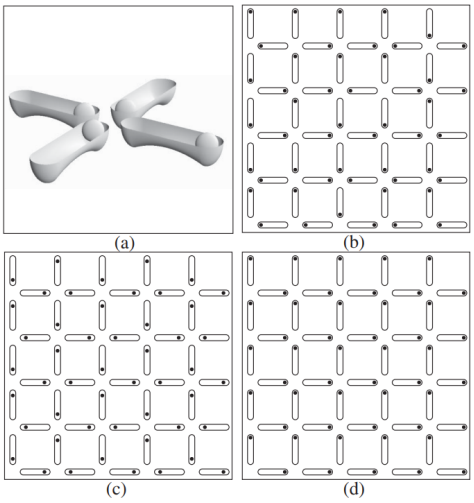

Figure 5: (a) Schematic of double well traps of colloidal ice unit cell. (b) Random distribution (c) Long range orderded ground state of square ice in the colloidal system (d) Biased system. Image by Libál et. al. [8].

Although promising, to make an experimental realization of the simulated system proved to be difficult. However, an alternative version of this system was first made by Ortiz-Ambriz and Tierno [9] where they featured paramagnetic colloidal particles trapped by gravity in a topographical array of double wells made with soft lithography. Traps were filled with colloids using optical tweezers. Repulsive interactions between colloids were induced by an external magnetic field perpendicular to the lattice plane. This magnetic field induces a parallel dipole moment in each particle

$$\mathbf{m} = \frac{\pi d^3 \chi}{6\mu}\mathbf{B}, \tag{2.3}$$

where d is the diameter of the particles, χ the volume susceptibility, and μ is the permeability of the medium (water). Contrary to ASI, dipole moments are out-of-plane, but an effective spin can be assigned pointing towards where the traps are filled, giving a different energy hierarchy to ASI; however, collectively the ice rule is enforced as magnetic interactions become stronger (see figure 6). Since thermal fluctuations are negligible due to the large size of the particles. The *cooling* process is not done thermally, but magnetically. The experiments started with a disordered process which can be considered as a thermal bath, while increasing the magnetic field strength induces an ordering to the system analogous to reducing the temperature.

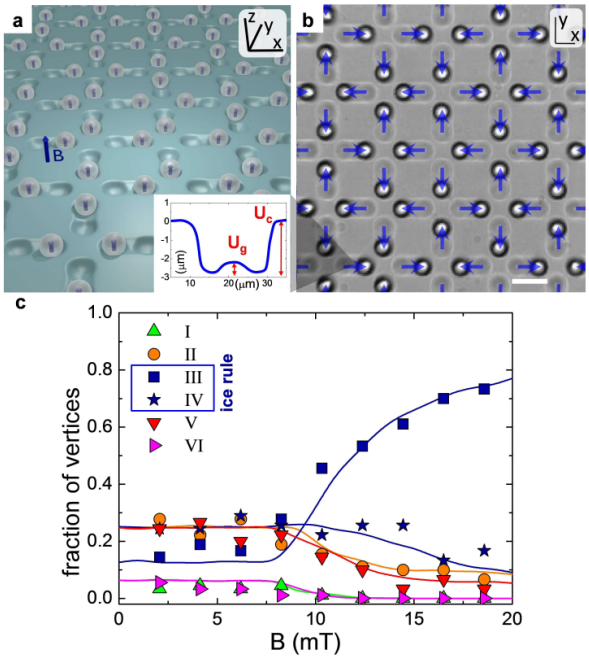

Figure 6: (a) Schematic of colloidal ice in a square lattice. Arrows represent the induced magnetic moment (b) Microscope image of the system. The arrows represent the associated spin. The scale is 20μm (c) Fraction of the vertices vs applied magnetic field. Scattered points represent experimental data, while lines represent simulations. Adapted image by Ortiz-Ambriz et. al. [22].

2.4 Relevant concepts on Phase Transitions and Criticality

Phase transitions are fundamental concepts in physics and materials science that describe abrupt changes in the physical properties of a substance as it undergoes a transition from one phase to another. These transitions typically occur as a function of temperature or pressure and are characterized by distinctive changes in macroscopic properties like density, magnetization, or specific heat. One of the most well-known phase transitions is the transition from a solid to a liquid (melting) or from a liquid to a gas (vaporization) as temperature increases.

At the heart of phase transitions lies the idea of an *order parameter* which is a measurable quantity that undergoes a sudden change as the transition occurs. The behavior of the order parameter helps classify phase transitions into different categories, such as first and second order phase transitions. *First order transitions* present a singularity in basic thermodynamic quantities such as the the particle density, or magnetization. *Continuous transitions*, do not present sudden changes in basic thermodynamic variables, thus the word *continuous*, but in second order derivatives of the free energy, hence these are commonly called *second order transitions*. Continuous transitions appear in phase diagrams at *critical points*

Near the critical point, certain thermodynamic properties like heat capacity, magnetic susceptibility, and correlation length exhibit divergence as the temperature approaches T_c. However, their behavior in proximity to the critical point is described by a set of parameters called *critical exponents*. These exponents describe the power-law relationships governing specific thermodynamic quantities as they approach the critical point, e. g. one expects the specific heat $C \sim t^{-\alpha}$ where $t = (T - T_c)/T_c$.

Landau theory is a powerful theoretical framework in condensed matter physics used to describe and understand phase transitions and the behavior of materials near critical points, it describes the functional

form of the free energy near criticality as a series expansion of the order parameter $\Psi(\mathbf{r})$ [25].

$$F = \int \mathrm{d}\mathbf{r} \left[A + Bh(\mathbf{r})\Psi(\mathbf{r}) + C\Psi^2(\mathbf{r}) + D\Psi^4(\mathbf{r}) + E(\nabla\Psi(\mathbf{r}))^2 + \cdots \right], \tag{2.4}$$

where $h(\mathbf{r})$ is a conjugated field, the constants $A, B, C, D, E, \ldots$ are temperature dependent, and F can be minimized by a proper choice of $\Psi(\mathbf{r})$.

2.4.1 The Glass Transition

The glass transition is a fundamental phenomenon in materials science and physics, marking the abrupt change in the physical properties of an amorphous material as it cools from a liquid state to a solid one, without undergoing a crystalline transformation. At a molecular level, it corresponds to the transformation from a highly disordered and mobile state to a more rigid, slow, glassy state. The ability of a material to form glassy states strongly depends on the time available for crystallization, and the mobility of its components.

A typical schematic on the glass transition is shown in figure 7a where the volume is monitored as a liquid is cooled slowly. A typical liquid-solid transition follows the liquid-crystal line where a discontinuous jump in volume is evident. However, transitioning to glass has more subtle changes and volume seems to change continuously for undercooled liquids. Although the nature of the glass transition seems a second-order phase transition, changes in the derivative, such as specific heat are not as sharp as in a true continuous transition (see figure 7b). Advances in the theory of the glass transition point toward a purely kinetic phenomenon strongly dependent on experimental time scale, rather than a thermodynamic phase transition since T_g marks the end in which the material exists in a metastable thermodynamic equilibrium [26].

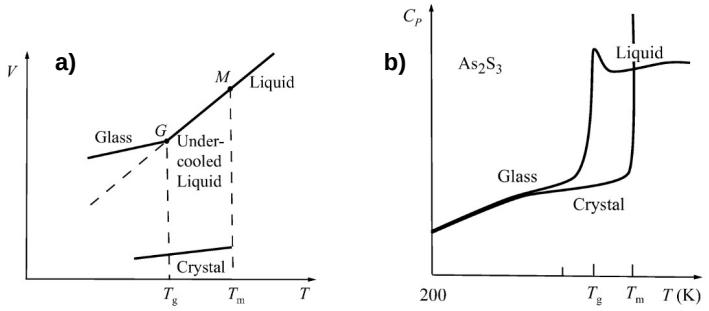

Figure 7: Schematic of the glass transition. (a) Schematic of volume as a function of the temperature of an undercooled liquid. (b) Specific heat of As_2S_3 near the glass transition. Edited figure from [2].

As mentioned in section 1.1, the glassy behaviour is commonly described in therms of a relaxation time τ_α greater than the experimental observation time which for colloids corresponds to browninan diffusion time (see equation 1.1). When working with the glass transition, a mobility field is introduced to characterize how much the particles move in a time frame [11]

$$c(\mathbf{r}, t, 0) = \sum_j c_j(t, 0)\delta(\mathbf{r} - \mathbf{r}_j), \tag{2.5}$$

where $c_j(t, 0)$ is a measure of how much particle j has moved between time zero and t, and the spatial average of this quantity describes how the system has evolved

$$\hat{C}(t, 0) = \frac{1}{V} \int \mathrm{d}r\, c(\mathbf{r}, t, 0). \tag{2.6}$$

Recalling the stochastic nature of Brownian dynamics, averaging time series over realizations of the experiments lead to the correlator

$$C(t) = \langle \hat{C}(t,0) \rangle_{\text{realizations}}, \tag{2.7}$$

that typically follows the Kohlrausch-Williams-Watts law (1.2) where τ_α is the relaxation time and b is a parameter which can be fitted. Further measurements include spatial correlation functions (1.3) that are expected to decay as (1.4) when the transition is approached.

3 Methodology

3.1 Molecular Dynamics and Brownian Motion

The main method employed in this study involves computer simulations, primarily relying on *icenumerics*, a Python wrapper designed to interface with LAMMPS (Large-scale Atomic/Molecular Massively Parallel Simulator). The wrapper was designed and is maintained by Dr. Antonio Ortiz Ambriz, the principal advisor for this research.

Simulations in icenumerics are done through the *magcolloids* package. In Molecular Dynamics, simulations are carried out by solving Newton's equations of motion

$$m_i \ddot{\mathbf{x}}_i = \sum_j \mathbf{F}_j, \tag{3.1}$$

for all particle and forces involved. In colloids, fluctuations in the liquid play an important role, hence the package has a modified version of LAMMPS to run Brownian Dynamics simulations. In Brownian Dynamics, it is assumed that all particles are immersed in a fluid where dissipation dominates, thus a drag force proportional to the velocity is included

$$\mathbf{F}_{i,\text{drag}} = -\gamma \dot{\mathbf{x}}_i. \tag{3.2}$$

Given that viscous forces are much greater that the inertial term in Newton's equation, an approximation can be made and $m_i \ddot{\mathbf{x}}_i$ can be ignored. Additionally, all particles are expected to be subjected to random interactions with the fluid, this can be included with a stochastic term in the equations of motion by adding a random variable η, also known as the Langevin thermostat, that has the following mean and correlations

$$\langle \eta \rangle = 0, \quad \langle \eta_i(t + \Delta t)\eta_j(t) \rangle = 2k_B T \gamma \delta(i-j)\delta(\Delta t). \tag{3.3}$$

Using these assumptions, the equations of motion are given as

$$\gamma \dot{\mathbf{x}}_i = \mathbf{F}_i + \eta, \tag{3.4}$$

and are solved numerically by the following discretization,

$$\mathbf{x}_{i+1} = \mathbf{x}_i + \frac{\Delta t}{\gamma}\mathbf{F}_i + \sqrt{2k_B T \Delta t \gamma} N[0,1], \tag{3.5}$$

where $N[0,1]$ is a normal distribution with zero mean and unit standard deviation.

3.2 Interactions

Particles (colloids) in the simulation are induced a magnetic moment via an external magnetic field, then there are two forces dominating the simulation:

- Trapping force.
- Magnetic dipole-dipole interaction.

The colloidal ice consists on colloidal particles confined in a bistable potential in such a way that colloids can jump from one hole to the other, mimicking a spin. The potential used in the simulations is a bi-harmonic potential of the form

$$\mathbf{F} = -kr_{\perp}\hat{\mathbf{e}}_{\perp} + F_{\parallel}\hat{\mathbf{e}}_{\parallel}, \tag{3.6}$$

here $\hat{\mathbf{e}}_{\perp}$ is a unit vector pointing away from the trap, k is the trap stiffness, and $\hat{\mathbf{e}}_{\parallel}$ is a unit vector pointing parallel to the trap. Here the orthogonal spring force prevents the particles from jumping to other traps. The parallel force $F_{\parallel}$ that confines the particles in either stable position is defined as

$$F_{\parallel} = \begin{cases} k(|r_{\parallel}| - d/2)\text{sign}(r_{\parallel}) & \text{if} \quad r_{\parallel} < d/2, \\ hr_{\parallel} & \text{if} \quad r_{\parallel} > d/2 \end{cases} \tag{3.7}$$

where h is the stiffness of the central hill, and d is the distance between the centers. This expression can be checked by allowing a particle to diffuse thermally through a trap, and observing its probability distribution.

Induced magnetic moments on the particles interact via the pairwise dipole-dipole potential energy given by;

$$U = -\frac{\mu m^2}{4\pi}\left[\frac{\mathbf{e}_i \cdot \mathbf{e}_j}{r^3} - \frac{3(\mathbf{e}_i \cdot \mathbf{r})(\mathbf{e}_j \cdot \mathbf{r})}{r^5}\right], \tag{3.8}$$

where $\mathbf{r}$ is the unit vector pointing along the line connecting the centers of the dipole moments. Then the force between dipoles is give

3.3 Metropolis algorithm

Given a certain set of conditions the system need to meet such as decimation or different external magnetic field, it is in our interest to determine the possible ground states or equilibrium states of such a system. To do this task a modified metropolis algorithm was implemented where at each Monte Carlo step the following operations are performed to propose a new colloidal ice configuration:

1. Flip one random colloid by inverting its direction and position relative to the center of the trap (figure 8a).
2. Flip four colloids in a randomly selected loop (figure 8b).
3. The energy of any configuration is given by pairwise dipole interactions from (3.8)

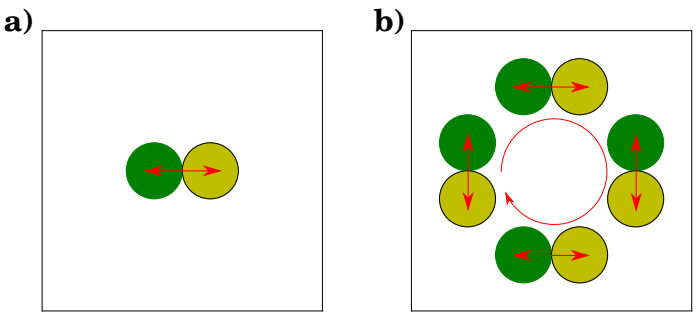

Figure 8: Procedure to generate new colloidal ice states. (a) One random colloidal ice is flipped. (b) Four random colloids in a loop are flipped.

3.4 Simulation Workflow and Analysis

The workflow for the simulation can be summarized in a few steps that are repeated several times. One of the most important aspects of colloidal ice is the experimental setup. Through the API of icenumerics all elements in the setup can be arranged in particular ways. We are interested in modifying several parameters such as: particle properties, traps properties, links in the graph (decimation), all features of the external magnetic field.

The main processes are stochastic, i. e. results from one simulation only correspond to fluctuations over the average. In order to have a more accurate representation of any physical behaviour, the simulation is ran several times (10-20 realizations) and any value comes from an average over the different realizations. All datasets are stored in `.csv` format from which the trajectories can be reconstructed and analyzed. Access to individual particle trajectories make it possible to compute correlation functions using built-in python functions.

The fraction of vertices of type i defined as follows

$$\nu_i = \frac{N_i}{N_{\text{total}}}, \tag{3.9}$$

where N_i is the number of vertices of a certain type, and N_{total} is the total amount of vertices. This is a commonly graphed quantity as a function of different parameters such as field strength, angle, time etc. This parameter gives a good idea on the energy hierarchy of the system, and whether a particular arrangement is preferred.

4 Workplan

The Master's program in Nanotechnology spans four semesters, and our approach to structuring the work plan is centered around setting specific goals for each semester (see figure 9). This method allows us to effectively guide our academic work, ensuring that we make the most of each stage in the program.

Activity	AD 2023	FJ 2024	AD 2024	FJ 2025
Topic exploration				
Introduction to icenumerics				
Preliminary simulations				
Literature reading				
Metropolis algorithm implementation				
Thesis proposal				
Programming and simulation				
Data analysis from simulation				
LAMMPS exploration				
Project for publication				
Publication manuscript				
Thesis writing				
Thesis defense				

Figure 9: Gantt chart in which the most important tasks to do during this research are covered.

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