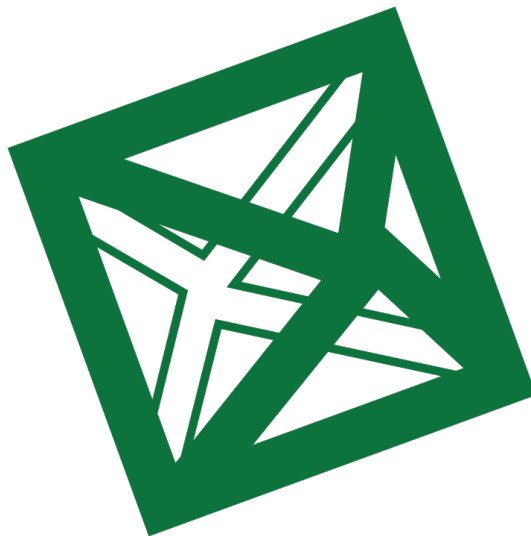


UNIVERSITÀ DEGLI STUDI DI  
MILANO-BICOCCA

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SCUOLA DI SCIENZE  
DIPARTIMENTO DI FISICA



Corso di Laurea Triennale in Fisica

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**Control of phase separating systems at criticality**

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**Relatore interno:**  
Francesco Montalenti

**Relatore esterno:**  
Olivier Pierre-Louis

**Candidato:**  
Rocco Suanno  
Mat: 858885

Seduta del 24/10/22

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Anno Accademico 2023–2024

## Abstract

A mixture of immiscible liquids or a binary alloy tends to separate into regions where each component is nearly pure, rather than remaining in a homogeneous mixture. These are examples of phase-separating systems. Even when examining the dynamics of a monolayer, which we will demonstrate is equivalent to a magnetic system (Ising model), the system undergoes phase separation under appropriate conditions. The system tends to leave the homogeneous state with zero magnetization (the unstable phase) and moves towards the stable states: all spins  $\uparrow$  or all spins  $\downarrow$  (the stable phases). This process, which does not require to overcome any energetic barrier, is known as spinodal decomposition and occurs when the temperature goes below a critical value  $T_C$ .

We study a continuous model that describes any system undergoing spinodal decomposition when the temperature is close to the critical value ( $T \simeq T_C$ ). This model is known as the Time-Dependent Ginzburg-Landau (TDGL) equation or the Allen-Cahn equation. To make our work more concrete, we will consider a specific system: an atomic monolayer interacting with a gas phase above it. However, the model has broader applicability.

We attempt to control the state of the system (position and shape of the magnetic domains) by varying the temperature over time. While we will not propose a specific strategy for varying the temperature, we aim to understand the properties of the state that can be controlled through temperature variations. Interestingly, we find that the primary feature of the dynamics, known as motion by curvature, is not influenced by temperature variations.

**Keywords**— Time dependent Ginzburg Landau, Allen-Chan equation, Control theory, Spinodal decomposition, Phase separation.

# Introduction

Controlling the morphology of a crystal's surface is a prominent challenge. The surface texture plays a crucial role in determining properties that have applications in various fields, such as regulating the growth of cell populations [2], enhancing the optical absorbance of materials [1], and producing nano-scale emitters with precise frequency outputs, like quantum dots [9]. The last application is relevant also in the semiconductor industry, where

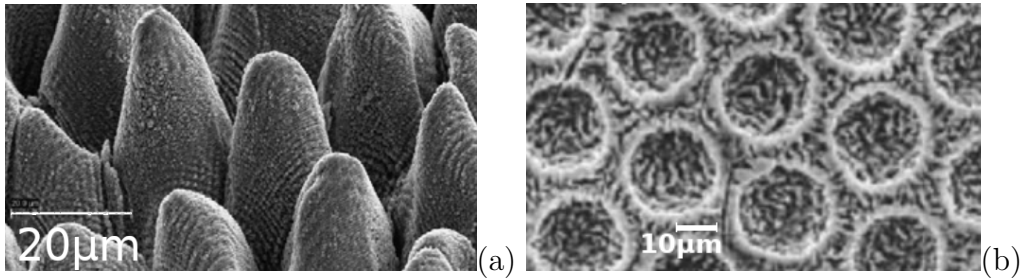


Figure 1: a) Laser treated Ti surface showing the formation of nano-structures after exposing the surface to approximately 450 laser shots with an intensity of  $0.9 \text{ J/cm}^2$  [1]. b) Ti surface with circular cavity of diameter  $d = 30 \mu\text{m}$ . Over the cavities, the surface presents an average roughness of  $0.7 \mu\text{m}$ . The circular cavities were prepared with photo-lithography, while the additional roughness was produced by post-treating the surface with an acid [2].

quantum dots are involved in the fabrication of nano-scale devices, like the single-electron transistor (Figure 2b).

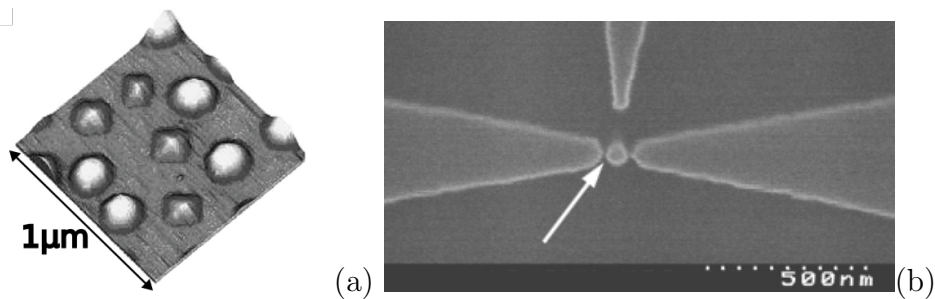


Figure 2: a) Atomic force microscope image of quantum dots produced by annealing a  $50 \text{ \AA}$  thick  $\text{Si}_{0.6}\text{Ge}_{0.4}$  layer at  $550^\circ\text{C}$  for 18 h [9]. b) Scanning electron microscope image of a single electron transistor. The arrow indicates the quantum dot [3].

Up to now, scientists mainly used *lithography* techniques to imprint a texture on the surface. This involves removing matter from the sample through a chemical compound, which reacts with the surface only where it is illuminated by light or a beam of electrons. But this method presents some limitations: the wavelength of the light limits the accuracy due to diffraction, while electron beam lithography requires expensive instruments. A new approach is represented by self-organized texturing. Here the system is brought out of equilibrium, for instance by some laser pulses [1]. Then matter reorganizes, reaching a new equilibrium state, that shows the presence of different nano-structures. Inspired by this idea, we studied how we can influence this morphology, keeping the system out of equilibrium by changing the temperature in time.

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In particular, we focused on the simpler case where the height of the sample cannot change more than one atomic layer, so we had to deal with a 2D system: a monolayer. Surprisingly, by making few natural assumptions, it's possible to show that this system is equivalent to the 2D Ising model, equipped with the Glauber dynamics, so the total magnetization is not conserved in time.

Then a fundamental aspect of this research, is that we didn't studied the dynamics of individual atoms. Instead, as we want to control nano-scale feature of the morphology, we adopted a continuum model. We choose the simpler model to describes a dynamics that minimizes the free energy, reflects the simmetries of the Ising model and where there aren't conserved quantities: the **Time Dependent Ginburg Landau equation** (TDGL), known also in mathematics as the Allen-Chan equation

$$\partial_t m(x, y, t) = \Delta m + C(T)m - m^3$$

where  $m(x, y)$  is the local averaged magnetization and  $C(T)$  is a parameter depending on the temperature.

The dynamics described by the TDGL equation has been extensively studied when the temperature is kept fixed in time [5][4][6] (Sec. ??), but in the literature there isn't any description of the dynamics following a variation of the temperature over time. The only paper that takes into account temperature variations [7] studies the effects of thermal noise, that is modeled by adding a stochastic term to the equation and neglecting any influence on the parameter  $C(T)$ .

In the past years, people also tried to *control* the state of the system, that means bringing the state closer and closer to a target state. The control was made possible by adding a source  $+f(x, y, t)$  in the equation, that is a function that you can arbitrarily change in time [8][10]. But no one considered the temperature as a force to drive the dynamics.

Our goal was to understand how the dynamics is affected by the modulation of temperature in time, and so what properties of the system can be controlled by varying the temperature in time. Surprisingly, the main feature of the dynamics is not affected at all by variations of temperature in time. This means that a modulation of temperature over time is not sufficient to control the main features of the state.

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# Chapter 1

## A simple model for the mono-layer dynamics

1.1 The Gas-Lattice model

1.2 The Landau-Ginzburg theory

1.3 A continuum model for the dynamics: the Time Dependent Ginzburg-Landau equation

## Chapter 2

# The Time Dependent Ginzburg-Landau equation (TDGL)

### 2.1 General introduction to the dynamics

### 2.2 1D dynamics

#### 2.2.1 Stationary states

#### 2.2.2 Interaction between kinks

#### 2.2.3 Evolution of an homogeneous (flat) state

#### 2.2.4 Stability of an homogeneous state: the condition for the formation of domains

### 2.3 2D dynamics

#### 2.3.1 Introducing a curvilinear coordinate system

#### 2.3.2 The principal mechanism of the dynamic: Motion by curvature

#### 2.3.3 Evolution of an isolated circular domain

### 2.4 Coarsening



# Chapter 3

## Controlling the dynamics by varying temperature over time

### 3.1 1D

#### 3.1.1 The adiabatic limit

#### 3.1.2 The leading order correction for slow varying temperature

### 3.2 2D

#### 3.2.1 When the temperature oscillates slowly in time

#### 3.2.2 When the temperature oscillates fast in time

## Chapter 4

# Controlling the typical size of domains by varying temperature over time

- 4.1 During the early dynamics
- 4.2 During the long time dynamics
- 4.3 During the intermediate dynamics

# Chapter 5

## Conclusions

### 5.1 Summary

### 5.2 Perspectives and applications

# Appendix A

## Computational tools

### A.1 Numerical methods for solving differential equations

#### A.1.1 Implicit and explicit schemes

#### A.1.2 The Cranck-Nicholson scheme

#### A.1.3 Example: TDGL equation for a flat profile

### A.2 Fourier transform

#### A.2.1 Discrete Fourier transform (DFT)

#### A.2.2 Spectral derivative

## Appendix B

# Simulating the Time Dependent Ginzburg-Landau equation's dynamics

B.1 The Cranck-Nicholson scheme in Fourier space

B.2 About the implementation in C

B.3 Choosing the simulation's parameters

# Appendix C

## Mathematical tools

### C.1 The Fredholm alternative

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