Collective Modes of Strongly-Correlated Electrons in Non-Translationally Invariant 2D Materials

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Abstract

Recent discoveries of charge density waves in Transition Metal Dichalcogenides, unconventional superconductivity in twisted bilayer graphene and other correlated phases of moiré materials, have raised questions about the role of quasiperiodicity in the enhancement of electronic correlations. Yet, these systems present a major challenge for theoretical modeling due to their lack of translational invariance. The goal of this proposal is to understand how correlated phases of strongly-correlated materials emerge in the absence of translational invariance and what is the role of quasiperiodicity and disorder effects in promoting correlated quantum phases of matter. We will focus on collective excitations which are experimentally accessible. We shall investigate how these collective modes impact fundamental properties of symmetry-broken states, namely superconductivity, magnetic and charge order in non-translational invariant two-dimensional materials. We expect our results to establish the role of moiré quasiperiodicity in promoting order and shed light on the mechanism underlying these phenomenona.

Contents

Chapter 1

Introduction

1.1 Two-dimensional materials

Since the experimental synthesis of graphene in 2004 by Novoselov *et al* [1], the field of two-dimensional (2D) materials has emerged as one of the most fertile areas in condensed matter physics. During the first decade, research activity in graphene expanded rapidly, with the number of publications growing exponentially, as illustrated in Fig. 1.1. For their pioneering contributions to this field, Geim and Novoselov, were awarded the Nobel Prize in Physics in 2010.

It was quickly understood that 2D crystals could be stacked to form new structures with completely different electronic properties [3]. Single layer graphene behaves as a semimetal whose conduction and valence bands touch linearly at the so-called Dirac points [4]. However, when two layers of graphene are stacked, the band structure changes, depending greatly on the particular stacking. For a bilayer system with a Bernal stacking [5] the bands touch quadratically and, in the presence of an electric field, there is a gap in the spectrum, dependent on the bias voltage [6]. This high tunability of the electronic structure may be then used for technologial applications.

Although carbon is a very stable element to chemical substitution, graphene is not immune to other forms of disorder, such as strain or adatoms and vacancies [7–9]. Disorder breaks translational invariance, precluding the use of the Bloch's theorem. In such cases, theoretical modeling of these materials becomes more complex, requiring a real-space approach. Transport properties are known to be suppressed in the presence of Anderson disorder (a random potential across the system) where the wavefunctions become exponentially localized. [10, 11]. In the next subsection, we will describe one interesting example of a perturbation that breaks translational invariance and is intrinsic and unavoidable in some 2D materials.
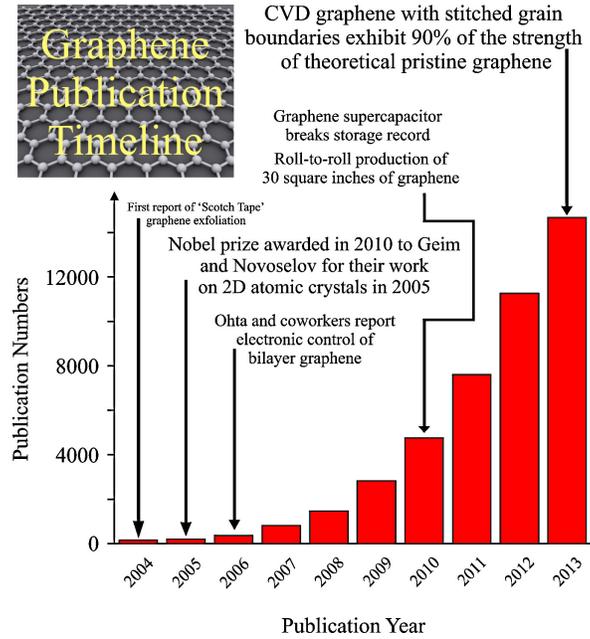
Figure 1.1: Number of publications that use graphene in the first ten years . The arrows mark some milestones. Taken from [2]

1.2 Moiré materials

It is known from wave theory that when two waves with similar frequencies interfere, a beating pattern appears. When two 2D Bravais lattices with different orientations (exhibiting some angle of rotation between each set of unit lattice vectors) or a mismatch in the lattice parameters, are stacked, they also interfere and form a Moiré pattern (see Fig. 1.2a)). The moiré pattern breaks the translational invariance, except for a discrete set of angles where it is possible to obtain a periodic structure [12] with a well defined unit cell. In condensed matter systems, moiré patterns trace back to the 80's where the last layer of graphite was found to have a random angle with the perfectly aligned bulk in Scanning Tunneling Microscopy measurements [13]. In 2007, Lopes dos Santos *et al.* provided a theoretical description for the electronic properties of bilayer graphene with a slight twist between the layers, coining the name twisted bilayer graphene (tBLG) [14]. Their main finding was the suppression of the Fermi velocity when compared to the single layer case, which was later confirmed in Scanning Tunneling Spectroscopy (STS) experiments. Twisted bilayer graphene samples with a random (and small) twist angle were studied [15] and it was found that two van Hove singularities appear near the Fermi level and, more interestingly, their energy separation decreases with decreasing twisting angle. Near a "magic angle", $\theta \approx 1.1°$, the two peaks could not be resolved. This was the discovery of the narrow (also known as flat bands) bands in tBLG. Some years later, Bistritzer and
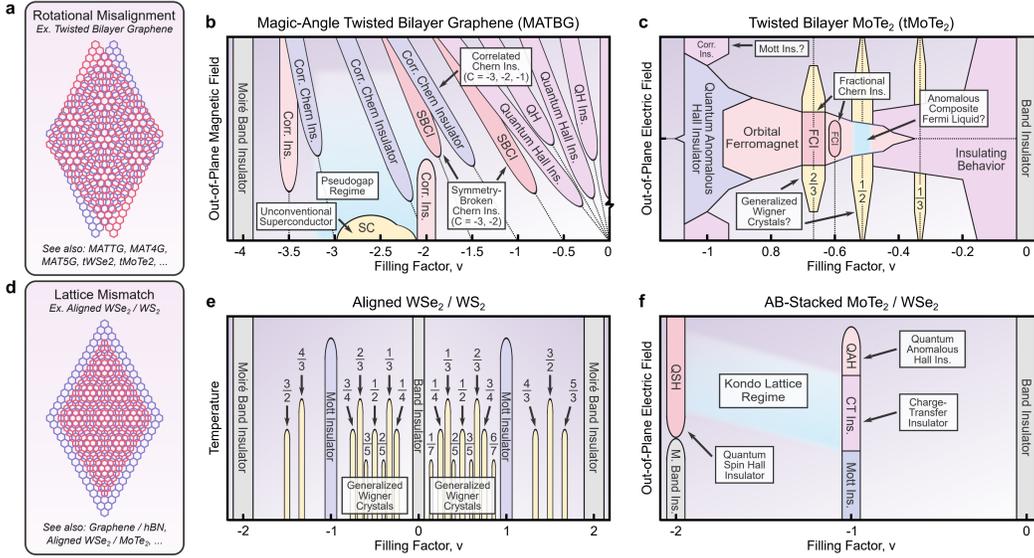
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Figure 1.2: Moiré materials overview: Lattice structure for a) Rotational Misalignment and d) Lattice mismatch. Phase diagram of b) Magic-angle twisted bilayer graphene, c) Twisted BilayerMoTe$_2$, e) Aligned bilayer of WSe$_2$/WS$_2$ and f) AB-stacked bilayer of MoTe$_2$/WSe$_2$. Taken from [37].

Macdonald, in their seminal work [16], proposed a new low-energy model and found out that the previously mentioned magic angle was the largest of a set of angles that lead to the suppression of the Fermi velocity. The presence of Van Hove singularities at the Fermi level increases the role of interaction effects, due to a magnification of the density of states, leading to novel correlated phases. However, the correlated phases were only observed in 2018, where Y. Cao, at the group of P. Jarilo-Herrero, was able to produce tBLG samples [17, 18] with a precise control over the twist angle, with the aid of the "tear-and-stack" technique. [19]. With electrical gates, they were able to vary the chemical potential and sweep the narrow bands and a full phase diagram of the low energy physics could be unraveled, encompassing superconductivity and correlated insulators. Today, the world of moiré materials has broaden and a whole set of distinct platforms has been used and intensively studied, either from a theoretical or experimental point of view. Examples that harness the rotational misalignment range from: i) twisted bilayer graphene [17, 18, 20, 21], ii) twisted double bilayer graphene [22, 23] , iii) twisted transition metal dichalcogenides (details in section 1.3) [24–29] to iv) other multilayer structures with multiple interfaces, such as quasicrystalline trilayer graphene [30] or even pentalayer graphene [31]. Another possibility to create a moiré pattern is by lattice mismatch in each layer, which is common in heterostructures such as aligned WSe$_2$/WS$_2$ [32, 33] or aligned MoTe$_2$/WSe$_2$ [34–36].

1.3   Transition Metal Dichalcogenides

Transition Metal Dichalcogenides (TMD) are another class of 2D materials belonging to the van der Walls material class. The chemical formula is MX_2, with M a transition metal (eg. Tungsten (W) or titanium (Ti)) and X being a chalcogen (eg. sulfur (S), selenium (Se) or tellerium (Te)). These type of materials have a layer-like shape where, in each layer, the atoms are tightly bounded through a covalent bound. Each layer can be stacked coupling through very weak van der Walls forces. These materials have drawn a great deal of attention due to their versatility in all realms of condensed matter physics. They exhibit exotic correlated phases such as unconventional superconductivity [38–43], Mott insulating phases [44] or charge density waves [38]. From a practical viewpoint, these materials are promising candidates in the study of collective modes being a possible mechanism for superconductivity, since they are more accessible to obtain experimentally than tBLG, for example. Despite the plethora of possible ground states, TMDs are also a great platform for the study of excited states such as excitons, tightly-bound pairs of electrons and holes formed due to the attractive Coulomb interaction. The optical properties of TMDs are usually dominated by the excitons and may be used for opto-electronical devices. In particular, composing different TMDs to form hetero-structures can lead to new emergent exciton states, such as interlayer [45, 46] or moiré excitons [47–50]. As stated above, disorder is unavoidable in 2D materials, and these materials are not an exception. The interplay between disorder and excitons alters the properties of excitonic response, being useful to technological approaches [51, 52].

Chapter 2

State of the art

2.1 Quasiperiodic moiré materials and correlated phases of matter

Moiré materials have emerged as the go-to platform to study correlated phases of matter, due to their remarkable tunability in twist angle and external parameters, such as electric and magnetic fields, temperature and carrier doping. A paradigmatic example is the aforementioned magic-angle twisted bilayer graphene, whose rich and notably complex phase diagram hosts a plethora of phenomena including unconventional superconductivity [18, 20, 21], correlated insulator phases [17, 20, 21, 53], a pseudogap regime under high magnetic fields [54], quantum Hall insulator phases [55], as shown in Fig. 1.2b). Some of these phases can be partially understood within a Hartree-Fock approach [56–61]. The dominant theoretical framework relies on a continuum model, which simplifies the system using a plane wave expansion that effectively turns the problem into a periodic one, exploiting the moiré length scale as the dominant scale [12, 16]. This approximation has been widely used due to its simplicity, specially for numerical works, and successfully captured some of the experimentally-observed results [62–64].

However, this description is only valid for a discrete set of angles, for which the system is exactly periodic with a well defined unit cell, allowing the application of Bloch's theorem. Outside this set of angles, the system is quasiperiodic (incommensurate) and translational invariance is lost. In these cases, Bloch's theorem no longer applies and the continuum model breaks down. To understand the implications of quasiperiodicity, it is instructive to revisit its effects in one-dimensional systems, before diving into quasiperiodic 2D moiré materials. In 1D, it is well established that a quasiperiodic potential, with an irrational ratio between the modulation wavelength and the lattice constant, can lead to localization transitions and the emergence of mobility edges [65, 66]. This is particularly noteworthy, since mobility edges in Anderson disordered systems only apear in three-dimensional systems.

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Quasiperiodicity, on the other hand, enables such phenomena to happen in one-dimensional systems. Furthermore, certain quasiperiodic modulations can give rise to multifractal wavefunctions, which are simultaneously delocalized in real and reciprocal space, rendering any effective continuum approach inadequate [67–72]. Building on this idea, Miguel Gonçalves *et al.* studied a realistic real-space model of twisted bilayer graphene near the magic angle, incorporating both periodic and quasiperiodic structures [73]. Remarkably, they found out that the flat band states become multifractal only in the quasiperiodic case, showing sub-ballistic transport, and being extended in both real and momentum space. In contrast, for periodic structures, the states remained ballistic and lacked multifractality. These findings underscore the limitations of the continuum approach and highlight the necessity of real-space methods in capturing the full physics of quasiperiodic moiré systems. This research proposal will be built around this idea.

From a theoretical perspective, adding interactions to quasiperiodic moiré materials remains largely unexplored. Investigating this interplay demands numerical methods capable of scaling to large system sizes of the order of 10^7 lattice sites. In a recent study [74], the authors analyzed a one-dimensional interacting model featuring flat-bands and multifractal states under a quasiperiodic modulation. They discovered a novel correlated phase arising exclusively in the regime with multifractal single-particle states. Crucially, this phase was absent when the modulation was periodic, despite the persistence of the flat band. This work demonstrates how multifractality, driven by quasiperiodicity, can fundamentally alter the many-body ground state. Further mean-field studies (made by the author of this research with a paper ready to be published in the following weeks), confirmed the emergence of the same phase, showcasing the potential of Hartree-Fock methods in this context. These findings suggest that multifractal states identified in quasiperiodic magic angle TBG may play a central role in shaping its correlated ground states. This hypothesis, yet to be fully explored in the literature, will be a central pivot in this research proposal.

Recent advances in experimental techniques have enabled the fabrication of twisted trilayer graphene with high precision. Each pair of layers forms a distinct moiré pattern, leading to the emergence of a new length scale, the superposition of the two moiré structures, see Fig. 2.1. This super-moiré length scale can be of the order of the micrometer [76]. Depending on the twist angle, these patterns can either be commensurate, forming a periodic superstructure, or incommensurate, leading to a moiré quasicrystal [77]. In experiments, it is possible to switch between the two regimes with a single device by adjusting the gate voltage [78]. At ultra low-energies, the system perceives only the super-moiré length scale and Bloch-like behavior persists. However, at accessible higher energies, the underlying quasiperiodicity becomes apparent at the moiré scale, justifying the term "moiré quasicrystal". Interestingly, in this regime, superconductivity is present on the phase diagram, indicating a link between quasiperiodicity and correlated phases. This opens a novel experimental setup to probe strongly interacting quasicrystals in a highly tunable and accessible setting, in contrast to traditional solid-state quasicrystals.
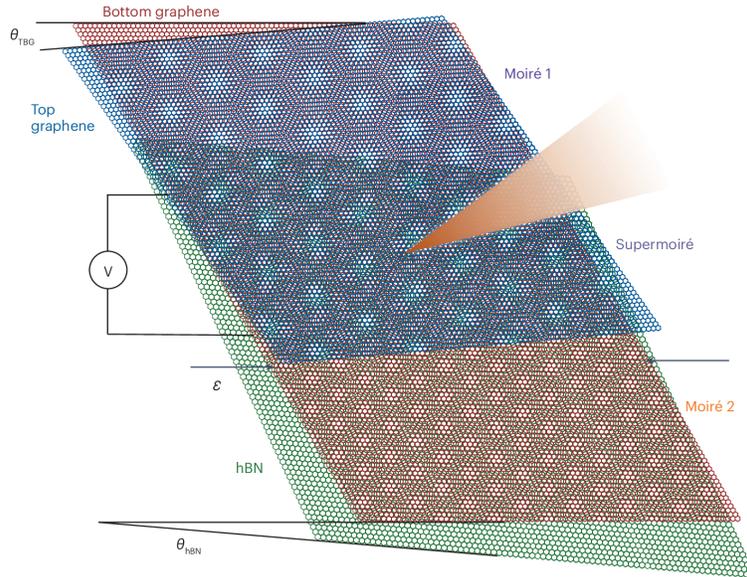
Figure 2.1: Schematic representation of a moiré quasicrystal. Taken from [75]

2.2 Collective modes

The mechanism behind superconductivity remains one of the most captivating and unresolved questions in condensed matter physics. The Bardeen-Cooper-Schrieffer (BCS) theory [79] provides a successful framework for understanding conventional superconductors, where electron-phonon coupling mediates an effective attractive interaction between electrons, enabling the formation of Cooper pairs and the emergence of superconductivity. However, a wide class of materials—collectively known as unconventional superconductors—do not fit within this paradigm. These include the cuprates (e.g., $YBa_2Cu_3O_{7-x}$), iron-based superconductors (e.g., $BaFe_2As_2$), and heavy fermion systems such as CeCoIn$_5$, where strong correlations and alternative pairing mechanisms are believed to play a central role. Among the most intriguing recent examples are the moiré materials, particularly twisted bilayer graphene, where the origin of superconductivity is still under active investigation. Remarkably, TBG exhibits a phase diagram that strikingly mirrors that of the cuprates, featuring superconducting domes flanking correlated insulating states. As such, unraveling the pairing mechanism in moiré systems may shed new light on the longstanding puzzle of high-T_c superconductivity, offering a promising route to deepen our understanding of strongly correlated electron systems. Many pairing mechanism have been proposed ranging from the strong electronic correlations [80–83], spin fluctuations [84] and topological textures such as skyrmions [85]. Other possible mechanism are collective modes, namely plasmons [86].

Nevertheless, modeling collective modes, especially in systems that lack translational symmetry, remains challenging. These approaches typically rely on Bloch's theorem [87], making it dificult to in-

corporate effects like disorder or quasiperiodicity. Addressing such effects demands a different strategy, such as computing fluctuations around a mean-field solution [88–90].

In other materials, such as TMDs, collective excitations like excitons, play a pivotal role in shaping the optical response. This further emphasizes the importance of incorporating collective effects into theoretical treatments of correlated systems.

Chapter 3

Research Proposal

3.1   Main Goals

The primary objective of this research plan is to investigate the properties of two-dimensional strongly correlated systems without translational invariance, specifically focusing on moiré materials such as twisted bilayer or trilayer graphene, and also Transition Metal Dichalcogenides (TMDs). As stated in chapter 2, the breaking of translational invariance, due to the presence a moiré pattern, strain or other forms of disorder, leads to new correlated phases of matter, and also on the collective modes that may arise due to interactions. This interplay has been ignored in the literature, and the main goal is to bridge this gap. To achieve this goal, we propose to develop simulation algorithms for strongly correlated electrons in real-space structures that accurately model these materials. This approach circumvents the limitations of commonly-used plane wave methods, which are not adapted to handle non-perturbative effects in structures lacking translational invariance. Our specific aims include:

1. Developing and benchmarking algorithms to study collective modes using a time-dependent Hartree-Fock method in real-space, suitable to tackle systems lacking translational invariance.

2. Computing the excitation spectrum and collective modes of tBLG and tTLG at the magic angle, comparing both periodic and quasiperiodic structures.

3. Investigating the role of collective modes as a mechanism for unconventional superconductivity in tBLG and tTLG, using Migdal-Eliashberg theory. Superconductivity was found to be enhanced in certain quasiperiodic systems with critical multifractal phases [91]. Here we will inspect if it can also be enhanced by quasiperiodicity in similar regimes for tBLG and tTLG.

4. Investigating the excitonic properties of TMDs and examining the presence of bound states, both with and without the charge density wave phase. The behavior of the collective modes, under the presence of the disorder and strain, will also be studied.

The outcomes of this project will contribute to a deeper understanding of collective modes in two-dimensional materials and their dependence on translational invariance. Our results have the potential to reveal novel mechanisms for superconductivity and other correlated phases with origin in inhomogeneities like disorder and quasiperiodicity. They will therefore have broad implications to a wide class of 2D materials where such features are ubiquitous.

3.2 Methodology

To tackle the previously described challenges, several methods will be employed:

3.2.1 Fermi Operator Expansion (FOE)

The ordered phases arising from strong correlations may be described by the reduced density matrix. Those phases can be computed using the Hartree-Fock method, which performs a variational search for the single-particle reduced density matrix that minimizes the ground-state energy. Using the fact that the reduced density matrix is "near-sighted" (decays exponentially for gapped systems and for gapless systems at finite temperature) [92] and combining it with a expansion in Chebyshev polynomials of the Fermi-Dirac operator [93], we can achieve a mean-field method that has a linear scaling with the system size, in contrast with the cubic scaling for the standard methods that involve the exact diagonalization of a single-particle Hamiltonian.

3.2.2 Time-Dependent Hartree-Fock (TDHF)

The time-dependent Hartree-Fock method is commonly employed in insulator contexts to determine exciton energy levels and their impact on optical responses. However, a formulation based on the reduced density matrix and its corrections through linear response theory enables the computation of generalized susceptibilities and instabilities of a mean-field symmetry-broken ground state. This method is essentially equivalent to solving the Bethe-Salpether equation for excitation energies, allowing the computation of sub-gap energy levels, such as collective modes (e.g., Goldstone or plasmon modes). Previously, this method was solely applied to moiré materials within continuum formulations, neglecting quasiperiodicity [94]. The method can be reformulated as an eigenvalue problem, with the corresponding matrix exhibiting unique symmetries. We will exploit those symmetries to reduce the scaling with system size [95].

3.2.3 Eliashberg theory of superconductivity

Superconductivity requires an attractive interaction between electrons. In the BCS theory, this is achieved by retardation effects due to the phonons, in the form of an electronic self-energy [96]. This

idea may be extended to other collective modes, for example plasmons. In both cases, the collective modes create a dynamical interaction that may be attractive, and the necessary pairing terms arise.

3.3   Work Plan

3.3.1   Mean-field ground states and many-body broken symmetries

(Months 1 - 9)

Using the FOE, we will obtain the Hartree-Fock ground state of quasiperiodic systems. First, to verify the methodology, we will consider simple generalizations of the paradigmatic Aubry-André, which exhibits exotic localization properties that we expect will directly affect the nature of correlated states. We will compare in detail the results for quasiperiodic and periodic structures with similar moiré lengths.

Then, we will proceed to obtain the ground state phase diagram of twisted bilayer and trilayer graphene in the temperature-doping plane (Milestone 1). To obtain results that are compatible with experiments, an extension to include superconductivity will be performed. The symmetry-broken states will be analyzed and compared with the experimentally known results. Also, differences between periodic and quasiperiodic structures will be characterized. We expect that the non-interacting sub-ballistic wave functions influence the phase diagram.

3.3.2   Collective excitations and instabilities of mean-field ground states

(Months 8 - 24)

The ground states calculated in Task 1 may contain zero-energy excitations known as Goldstone modes. Using the Time-dependent Hartree-Fock, we will study the excitations, therefore the collective modes, to check for those instabilities. We will start with the 1D model, to verify the methodology and obtain a complete understanding on how to interpret the collective modes. With the method well established, we will compute the collective modes on quasiperiodic twisted bilayer and trilayer graphene (Milestone 2). The possible different collective modes will be analyzed. Experimentally accessible response functions, like the optical conductivity and charge and spin susceptibilities, will be computed including these sub-gap excitations [97]. We will compare these response functions for periodic and quasiperiodic structures to understand the impact of the lack of translational invariance.

3.3.3   The role of collective modes on the unconventional superconductivity

(Months 21 - 36)

With the computed collective modes, we will study possible mechanisms behind the unconventional superconductivity in twisted bilayer and trilayer graphene (Milestone 3). We will employ a variation

of the Migdal-Eliashberg theory that considers other collective modes, such as plasmons, instead of the phonon-mediated attractive interaction.

3.3.4 Interplay between disorder and excitons in TMDs

(Months 37 - 42)

Our implementation of the TDHF method will enable the calculation of excitonic bound states in TMDs and other 2D insulators, accounting for disorder effects, as opposed to conventional reciprocal space approaches (Milestone 4). We will calculate optical conductivities and magnetic response functions using realistic models and compare them with experimental findings.

3.4 Mitigation Plan

If there are any limitations with intensive numerical computations, we will explore alternative formulations of the continuum models, which are less computationally demanding, to check wether it is still possible to introduce quasiperiodicity. For one-dimensional systems, we may also utilize the Density Matrix Renormalization Group [98], a well-estabilished method for computing interacting ground states.

3.5 Computational infrastructure

The most important part of this research proposal will be the use of High Performance Computing (HPC). The author of this research proposal is associated to the Center of Physics of Universities of Porto and Minho, therefore having unlimited access to the Cluster Gird at FEUP. Additionally, the author is already engaged in a project, hosted by the National Network for Advanced Computing (RNCA), in which he is a Co-Principal Investigator. This project has been allocated roughly 1.000.000 computational hours over 12 months, in the Deucalion Cluster, under the reference 2024.09396.CPCA.A2. This allocation underscores the scope and potential of this current research. Additional grants are expected in the future.

3.6   Timeline

	Task		Year 1												Year 2												Year 3												Year 4											
		1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19	20	21	22	23	24	25	26	27	28	29	30	31	32	33	34	35	36	37	38	39	40	41	42	43	44	45	46	47	48
1	Fermi Operator Expansion																																																	
2	Time-dependent Hartree-Fock																																																	
3	Eliashberg theory of Superconductivity																																																	
4	Excitonic responses on 2D disordered systems																																																	
5	Finishing Thesis Writting and Submitting																																																	

All tasks will be performed at
Centro de Física do Porto
(CF-UM-UP)

M1   M2   M3   M4

Milestones:

M1	Phase diagram on Twisted Bilayer / Trilayer Graphene on Temperature-Dopin Plane
M2	Obtaining the collective modes of twisted bilayer / trilayer graphene and the role of quasiperiodicity
M3	Understanding the effect of collective modes on the mechanism of superconductivy on twisted bilayer / trilayer graphene
M1	Obtaining the effect of disorder on the excitonic bound states of TMDs

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