

ERC Advanced Grant 2018
Research proposal [Part B2]
(not evaluated in Step 1)

Part B2: The scientific proposal (max. 15 pages, references do not count towards the page limits)

Section a. State-of-the-art and objectives

A.1 Introduction.

Some of the most fascinating open questions in science have to do with the emergence of non-trivial properties when small quantum objects, that we understand very well, assemble to form larger structures [Anderson72]. Dramatic examples of emergence are crystals that display superconductivity or Quantum Hall effect, and biomolecules that replicate themselves, giving rise to life. The ultimate origin of the particles that we now consider as elementary, and even the quantum laws that we use to describe them, might also be an emergent property of smaller microscopic constituents. The depth and breadth of quantum emergence permeates a major fraction of research areas in physics, chemistry and biology.

Conventional computational strategies to address emergence in quantum systems face the so called exponential wall problem [KOH99]. The computational resources needed to model a quantum system in a classical computer scale exponentially with the number of degrees of freedom. Therefore, exact numerical solutions of the many-body problem are only possible for very small systems. As a result, the entire business of condensed matter theory is devoted to creative approximations and heuristic approaches, with a record that combines great successes, such as the understanding of fractional quantum Hall effect [LAU83], one dimensional spin systems [HAL18, WHI92] and superconductivity in conventional metals [BCS57], but also great failures, such as the lack of a theory for high temperature superconductors.

The exponential wall is also a severe problem in quantum chemistry, and hampers progress in computer assisted drug design, in our understanding of biomolecules used by nature to efficiently solve problems, such as nitrogen fixation and photovoltaic conversion, outperforming our best current technologies. Finding a viable universal approach to solve the many-body problem, going around the exponential wall, would enable tremendous progress in many areas of science, going beyond condensed matter physics, and would also have a strong technological impact.

Quantum simulation was proposed by Feynman [FEY82] to explore quantum systems circumventing the exponential wall. Broadly speaking, quantum simulation can be carried with two complementary approaches [BUL09,GAI09,HAU12,CIR12,ACI18]. First, the so called analog quantum simulation (AQS), the properties of model Hamiltonians are explored by building quantum systems engineered to provide a physical realization of these models. Second, there are quantum algorithms [LLO96] that run on universal quantum computers and permit to compute exactly the ground state energy and other relevant physical quantities [WEC15] in the many-body problem. This approach is known as Digital Quantum Simulation. In both instances, quantum simulation dilutes the distinction between computation and experiment and, more importantly, goes around the exponential wall problem by using quantum resources, such as entanglement and coherence, to model quantum systems.

The implementation of quantum simulations, either digital or analog, requires the fabrication of artificial quantum lattices: engineered arrays of quantum objects whose interactions can be modulated in order to produce non-trivial emergent properties and/or functionality. The most advanced type of artificial quantum lattice are quantum computers.

A.2 General Objectives

This proposal explores solid state based strategies to create new artificial quantum lattices to carry out Analog Quantum Simulations that permit to solve the quantum many body problem. More specifically, I propose to break ground on three outstanding problems that build on a combination of recent experimental breakthroughs and my own specific expertise:

Objective 1: To design a platform for analog quantum simulation of Hubbard model that permits to explore a wide range of lattices, filling factors and temperatures. For this matter I propose to use graphene bilayers with an ordered array of sp^3 defects that introduce localized electronic states within the

gap. This permits the simulation of the Hubbard model in a variety of artificial lattices with tunable on-site Coulomb repulsion, hopping energies and filling factors, in the quantum degenerate limit. The proposal addresses strategies for fabrication and validation of such a system, that will enable analog quantum simulations of a variety of non-trivial electronic phenomena, including unconventional superconductivity[AND97], quantum spin liquids and quantized anomalous Hall effect[MAR08].

Objective 2: To design a platform for analog simulation of spin systems with short range interactions, based on engineered adatom arrays on surfaces. Here I propose to take advantage of arrays of magnetic atoms nanoengineered on surfaces using scanning tunneling microscopes (STM), as a platform for AQS of spin Hamiltonians. The focus will be placed on the simulation of quantum disordered phases such as spin liquids in 1D and 2D. This should permit a radically new way to explore quantum magnetism with artificial lattices.

Objective 3: To find an efficient system to implement time-controlled laser induced exchange interactions between local spins in a semiconductor. Optically induced exchange interactions (OEI) hold the promise[PIER02] of exchange interactions between distant local spins that can be switched on-and off with laser pulses. The goal here is to carry out a systematic computational study of the right combination of host material and local spins to make this happen experimentally. For the host semiconductor I will exploit the very suitable optical properties of two dimensional transition metal dichalcogenides, that include very large Rabi coupling [LIU15], and very large excitonic binding energy [WAN18], in order to enhance the magnitude and the range of the OEI. For the local spins, I will consider a variety of point defects, molecular adsorbates, nuclear spins and dopants.

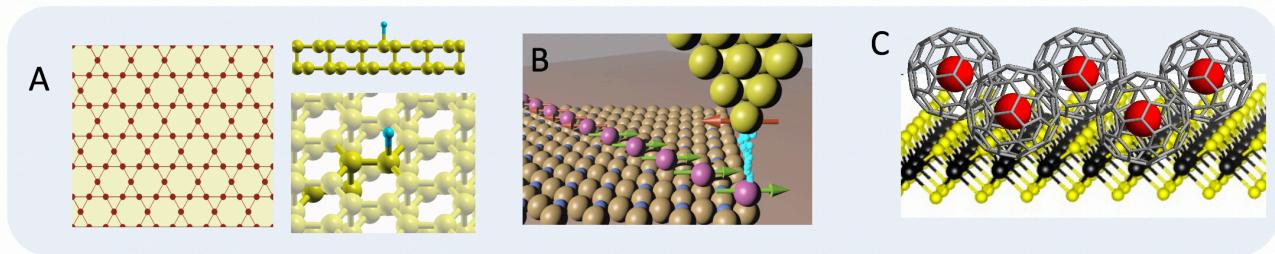


Figure 1 Artificial quantum lattices. **A:** Kagome lattice of localized electron states created around carbon sites passivated with atomic hydrogen in Graphene bilayer **B:** a chain of magnetic atoms on a metallic surface, probed and positioned with a STM.. **C** Array of paramagnetic centers ($N@C_{60}$) adsorbed on a transition metal dichalcogenides. Exchange interactions are induced by optical subgap radiation.

A.3 Timeliness

The developments that make this proposal particularly timely are the following:

- 1) The demonstration of atomic scale positioning of atomic hydrogen on graphene [GOL16] and the verification of my theoretical work [PAL0,SOR10] showing that this hydrogen functionalization induces a weakly localized state, occupied with a single electron, and extended over a few nanometers. The experiment [GOL16] shows that it is possible to create an array of these localized states with atomic scale precision.
- 2) The fabrication of functional structures where thousands of atoms are positioned, with atomic precision, using STM [KAL16], with my participation in the theory part.
- 3) The invention of two experimental techniques, STM- inelastic electrons spin spectroscopy [HIR06, JFR09] and STM-electron spin resonance [BAU15,FER17] that permit to probe and manipulate individual atomic spins with exquisite resolution, including the recent observation of hyperfine spin splitting [WIL18b] with my participation in the theory part.
- 4) The discovery of semiconductor two dimensional materials that have a very large coupling to light [LIU15] and can be functionalized in several ways. This makes it possible to envision arrays of localized spins controlled by laser induced spin interactions.

The choice of material systems for this proposal has been fully informed by the current state of the art. My strong background of collaborations with top experimental groups in the area of 2D materials [KLE18] and STM [OBE14,SPI14, KAL16, YAN17,WIL18,WIL18b] motivates the choice of systems and approaches in order to maximize the feasibility of the predictions that QUASAL will

produce. Success of this proposal will permit dramatic breakthroughs in our understanding of quantum matter.

A.3 State of the Art and Specific Objectives

In this section I review the state of the art relevant for the proposal and I breakdown the 3 general objectives in a list of more specific objectives.

A.3.1 Analog quantum simulation of model Hamiltonians.

Model Hamiltonians, such as the Hubbard model for electrons and the Ising and Heisenberg models for spins, play a central role in our understanding of quantum matter for the following reasons:

- They can be formulated in a very simple way, yet their solutions, either approximate or exact, describe physical phenomena that occur in real materials, such as the Mott-Hubbard metal-insulator transitions, broken symmetry states, etc. The Hubbard model is believed to describe the mechanism for high temperature superconductivity [AND97].
- Both the Heisenberg and Hubbard model Hamiltonians have exact solutions in 1D and rigorous theorems, such as the Lieb theorem[LIE89] for bipartite lattices, that serve to benchmark numerical methods.
- New paradigms, such as topological quantum matter[HAL18], fractional excitations, and spin-charge separation,, and new calculation methods, such as density matrix renormalization group (DMRG), have been discovered using Heisenberg and Hubbard models.

There are no efficient methods to solve the Hubbard and Heisenberg models, and their variants, once we venture out of the 1D case. As a result, there are several open outstanding questions that remain unanswered, such as the origin of high T_c superconductivity in doped Mott insulators, initially observed in cuprates and recently found in twisted bilayer graphene as well [CAO18b]. Standard numerical calculations can only deal with embarrassingly small systems. For instance, state of the art papers report diagonalization of Heisenberg model with up N=50 S=1/2 sites [WEI18]. This reflects the extreme inefficiency of classical computers to store quantum information and, as anticipated by Feynman, calls for a quantum strategy to model quantum systems[FEY82].

A quantum simulator, as defined by Eisert et al. (in chapter 4 in [ACI18]) is “*any physical quantum system precisely prepared or manipulated in a way aimed at learning interesting properties of (...) quantum systems*”. Making a working quantum simulator is an extremely challenging task. Here I want to build the case for the design of new quantum simulating platforms that will provide a dramatic leap forward in our understanding of flagship model Hamiltonians.

The 3 main objectives of the proposal deal with solid state platforms for Analog Quantum Simulation of model Hamiltonians [BUL09, HAU12,GEO14,AC18]. This needs a physical system whose effective Hamiltonian is governed exactly by the model Hamiltonian to be simulated. At this point, the most advanced platforms for AQS are cold atoms and trapped ions. Therefore, it would be particularly important to develop new solid-state based platforms that outperform the existing AQS platforms and can be integrated with conventional electronics.

A.3.2 Analog quantum simulation of Hubbard models.

Several platforms have been used to model the Hubbard model. Most of them are static simulators of the equilibrium properties, which are of great interest in condensed matter. All of them are still far away from providing faithful simulations in the interesting properties of the model. For instance, cold atoms, both bosonic and fermionic, have been trapped in optical lattices with various geometries (square [COC16], honeycomb[UEH13], Kagome [JO12]). In addition, the ratio of U/t can be changed significantly. However, these experiments can not explore the limit of temperature much smaller than the hopping and repulsion energies, where most of the interesting quantum emergent phenomena take place. For instance, in reference [COC16], temperature is $k_B T > 0.6 t$, and therefore, much bigger than $J=t^2/U$, the energy scale for super-exchange spin-spin interactions.

The simulation of Hubbard models with solid state platforms permits to build systems with much larger t and U, so that the $k_B T \ll t^2/U$. Attempts to do this have been carried out using two different strategies. In [SIN11], authors trapped electrons, confined as a two-dimensional gas in a gallium arsenide quantum well, in a nanofabricated lattice with honeycomb geometry, providing a platform to realize artificial graphene. Indirect evidence of the formation of a Hubbard model was inferred from Raman spectroscopy of the collective excitations, showing new collective modes. However, there are several issues with this approach. First, the role of disorder fluctuations, uncontrolled fluctuations in the patterning, that could result in fluctuations of the

confining potential. Given the size of the effective Hubbard U, estimated to be 2 meV, this could be an issue. Another problem in this approach is the control of U/t. In this platform, the extension of the wave function of the localized states was changed by application of a perpendicular magnetic field. This method is not optimal, as this also couples to the spins. After 7 years since this seminal experiment, there are very few follow up experiments[NAD12,WAN16], which suggests that this approach has issues that call for an alternative.

A stronger confinement can be obtained using either quantum dots [HEN17] or single dopants[SAL16]. However, in both cases, the number of Hubbard sites is very small (N=2 for the dopants, N=3 for the dots). As a trade off, the hopping is in the range of 0.3 meV, that is 50 times larger than the temperature, and the system is in the strong coupling regime ($U = 7 t$). The fabrication of a controlled array of 3*3 quantum dots has been reported in August 2018, showing where do we stand in this type of platform.

At this point there is no platform that permits to simulate the Hubbard model, and has at the same time all the desirable features: 1) tunable U/t ; 2) a number of sites large enough as to make classical simulation impossible (>40); 3) relevant energy scales ten times larger than temperature; 4) a wide tunability of the carrier density. In this proposal I propose a radically new approach and I intend to show that functionalized graphene bilayers meet these 4 requirements.

A.3.3 Simulating Hubbard models in Functionalized Graphene bilayer.

Objective 1 of this proposal is to implement an analog quantum simulator of the Hubbard model taking advantage of the unique properties of graphene bilayers (GBL). GBL have most of the interesting properties that make graphene monolayers so interesting and attractive, including very high mobility and the same potential for functionalization and carrier density control by gating. In addition, graphene bilayers have two more properties absent, in graphene monolayers, that I exploit in this proposal:

1. Application of an off-plane electric field opens up a gap, from zero up to 250 meV [ZHA09].
2. Correlated electron physics, including exotic superconductivity, has been observed in twisted bilayer graphene, linked to the emergence of flat bands associated to a triangular array of localized electronic states [CAO18a,CAO19b]

The radically new approach proposed here is to choose the location of these localized electron states by implanting them by means of sp^3 functionalization controlled with atomic scale precision using STM. In this type of functionalization, the chemisorbed object and the p_z orbital of one carbon atom of a graphene layer form a strong covalent bond. As a result, both the p_z orbital and 1 electron are removed from the extended covalent network formed by graphene. This results in one unpaired electron which entails the formation of a localized state at the Dirac point. The simplest and most celebrated example is functionalization by chemisorption of atomic hydrogen[YAZ07], but many others are possible[SAN12]. The properties of chemisorbed atomic hydrogen on graphene been widely explored theoretically [YAN07,PAL08,SAN12,GAR17] and probed experimentally, using scanning tunneling microscope (STM) spectroscopy [GON16].

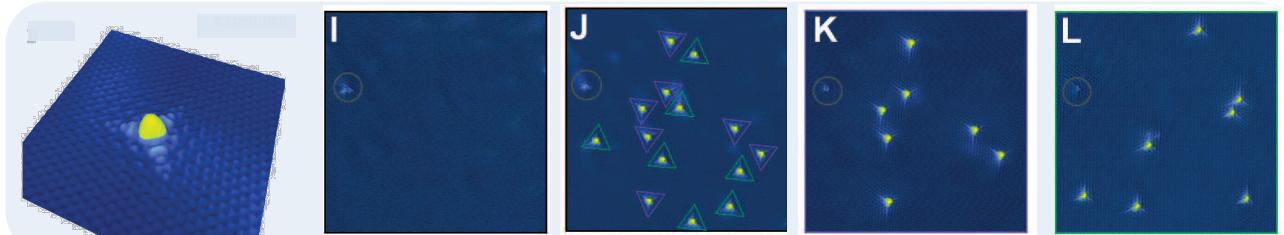


Figure 2 Atomic scale manipulation of Hydrogen on graphene, taken from [GON16]. **A:** STM scan of a single hydrogen atom on graphene ($7 \times 7 nm^2$). **I-J-K-J:** 4 steps of a manipulation sequence, all in the same region of ($28 \times 28 nm^2$). **I:** Pristine area (notice a point defect). **J:** 14 hydrogen atoms are placed, equally shared by the 2 sublattices. **K** and **L:** Only atoms of 1 sublattice are preserved, the others selectively removed.

The atomically accurate positioning of the binding site of hydrogen atoms on graphene has been demonstrated experimentally [GON16]. As shown in figure 2, González et al [GON16] managed to move hydrogen atoms over the surface, and to selectively place up to 7 hydrogen atoms in the same sublattice. In private communication, the leading author of this experiment, Ivan Brihuega, has confirmed that atomic manipulation could be achieved, in principle, with a much larger number of hydrogen atoms. The STM manipulation of more than 8.000 atoms has been demonstrated using other platforms, such as chlorine atoms on Cu(100) [KAL16] or CO₂ molecules on Cu(111) [GOM12].

The sp^3 functionalization of graphene bilayer is also expected to create a localized state that hosts 1 electron with energy close to the Dirac point [NIL07]. Application of an electric field perpendicular to the graphene opens up a gap. Using a dual gate, the carrier density and the band-gap can be changed independently [TAY10]. In particular, the band gap can be open without adding electrons or holes into the bands. Preliminary calculations carried out in my group [GAR18] show that, for a wide range of fields, the energy of the sp^3 localized state stays inside the gap (see figure 3), hosting a single electron.

Importantly, the extension of the localized state changes radically with the applied field (see figure 3). At zero field, there is no gap and the state is a resonance, with an infinitely extended wave function. As the gap opens, the state becomes a normalized bound state, that can be shrunk down to a few nanometers. The modulation of the extension of the state changes two important energy scales: hopping with adjacent states and the Coulomb repulsion associated to the addition of a second electron into the state, the Hubbard U . More localized states have a larger addition energy U and smaller hopping t .

A dimer of sp^3 defects defines a two site Hubbard model whose relevant energy scales can be tuned with two independent parameters: the distance between the functionalized sites and the extension of the states. Thus, for a given dimer distance, the electric field can control the transition between weak coupling (extended states, large hopping, small U), and strong coupling (localized single particle states, small hopping, large U).

Preliminary calculations carried out in my group [GAR18] show that ordered arrays of sp^3 functionalizing centers defines crystals with new sets of bands, including a new set of in-gap bands associated to the sp^3 induced states. We have carried out calculations for 3 lattices, triangular, honeycomb and Kagome [GAR18]. Results for an artificial honeycomb crystal where the sp^3 centers are 8.4 nm apart, with tens of thousands of atoms, are shown in figure 3C, for two different values of the off-plane electric field. We are able to fit the two in-gap bands (blue, barely visible due to high quality fitting) with a model of a honeycomb lattice with 1 orbital per site and up to third neighbor hopping (orange). This shows, conclusively, that this system realizes an artificial graphene, with a narrow bandwidth (20 meV) that can be tuned electrically.

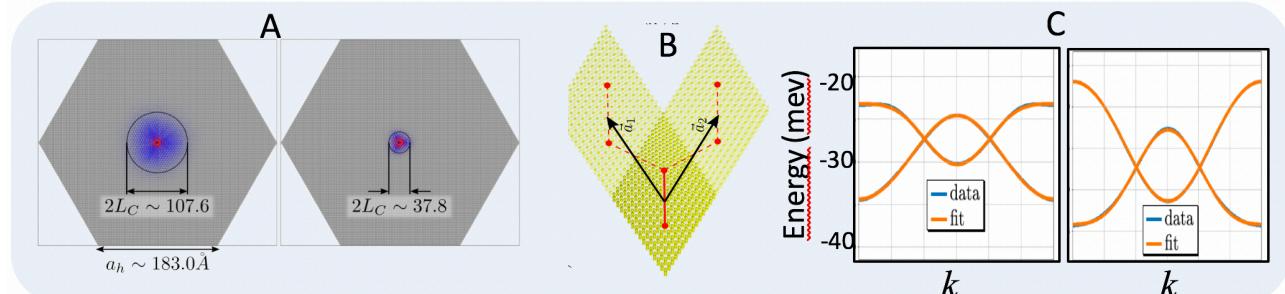


Figure 3: Electric control of hopping in Hubbard model simulator. **A.** A map of the wave function for a single sp^3 , for two different values of the electric field. **B.** The unit cell of a graphene bilayer functionalized with an array of sp^3 defects, 8.4 nm apart, that form a honeycomb pattern. **C.** In-gap bands, corresponding to B, for 2 different values of the electric field (blue) fitted to a simple tight-binding model (orange).

Our preliminary results [GAR18] (see Fig. 3) show the potential of this functionalized graphene bilayer platform to simulate the Hubbard model in a variety of systems, including finite size planar arrays, chains, and 2D lattices. The strength of the Coulomb repulsion U depends on the extension of the impurity states, that is controlled electrically. In addition, the filling factor of these states can also be controlled independently, using a dual gate [TAI10]. Therefore, this platform permits in principle to explore Hubbard models changing the lattice geometry, the t/U ratio and the filling factor, providing therefore an extraordinarily powerful tool to explore correlated electronic phases. Including superconductivity in a square lattice [AND98], a Chern phase in the triangular lattice at $\frac{3}{4}$ filling [MAR08], and to elucidate the nature of the spin liquid phase of the triangular and Kagome lattices for the AF Heisenberg model, that is obtained in the strong coupling $U>>t$ limit of the Hubbard model.

An obvious challenge to implement this idea is the manipulation of hundreds of hydrogen adatoms on the surface of graphene bilayer using STM manipulation. Although the fabrication of artificial lattices where the position of up to eight thousand atoms is determined with atomic scale precision has been demonstrated [KAL16], I will explore an additional route to fabricate the arrays: the formation of self assembled monolayers (SAM) of planar radical molecules deposited on top of the graphene bilayer. The formation of an ordered array of spinful molecules on top of graphene has been demonstrated experimentally [GAR16]. In this proposal I will explore the electronic properties of graphene bilayers functionalized with very large planar molecules that induce a sp^3 functionalization of graphene.

Summing up, the list of specific **tasks** to reach the 1st objective of having are:

- 1) **Mapping:** To compute the effective interacting fermion Hamiltonian that describes the in-gap states of the functionalized graphene bilayer, including the hopping matrix, the Hubbard coupling, as well as other many-body coupling that might also be present.
- 2) **Self assembly:** to model, using density functional first principles calculations, the electronic structure of graphene bilayer functionalized with large planar radical molecules, to assess their viability to act as large area sp³ centers.
- 3) **Probing:** to model electron transport, both in-plane and STM, and to relate the dI/dV curves to relevant spectral functions of the Hubbard model. This is essential to provide a connection between experiments on the simulating platform and the Hubbard model.
- 4) **Validation:** Validation of the simulating platform involves the independent verification of the simulation results. This is only possible when alternative simulation methods are available. For that matter, the simulation of small size cluster and 1D chains of the Hubbard model, will be taken as bench-marks.

A.3.4 Analog Quantum Simulation of spin models: exiting platforms

Analog quantum simulations of spin models has been studied with several platforms such as trapped ions[POR04] and superconducting qubits[DAL15]. I claim that the experiments with engineered magnetic adatoms are also a platform for quantum simulations of equilibrium properties and this proposal intends to fully exploit this possibility. The use of magnetic adatoms is discussed in detail in the next sections. Here I focus on the existing platforms.

The use of trapped ions to do quantum simulations of anisotropic Heisenberg (XXZ) model was proposed by Porras and Cirac [POR04]. They showed that this type of platform permits to induce spin-spin coupling between distant ions, both for electronic or nuclear spins, by driving the spins with spin-dependent optically induced forces. This platform is very convenient for several reasons. First, ion spins have very long coherence times, which also motivated the proposal to use them as a platform for universal quantum computing[CIR95]. Second, individual ion spins can be addressed [WIN87], with single shot optical readout. Third, ions can be trapped in arrays of various geometries, including linear arrays in linear Paul traps [SEN15, ZHAN17] as well as 2D lattices in Penning traps[BRI12]. Fourth, the number of trapped ions whose interactions can be controlled can go above 50 for linear traps and more than 200 hundreds in 2D arrays formed in Penning traps[BRIT12]. Fifth, although most of the work has focused on S=1/2 systems, the simulation of S=1 systems has also been demonstrated in linear traps[SEN15]

Although ion traps constitute a truly remarkable platform for quantum simulation of spin systems, they have a few limitations that motivate my quest for complementary strategies in the second objective of this proposal. The first and more important limitation is the long-range nature of the effective spin-spin coupling, that decays with distance as a power law $J(r) = r^{-\alpha}$, with a ranging between 0.5 and 1.5. This prevents the quantum simulation of spin models with short range interactions, relevant to describe solid state materials. Second, with the state of the art, ion traps only include one type of ion, although this could be worked out in principle. Here I propose a radically new approach to go around these two problems: the use of nano-engineered magnetic adatoms as a platform for quantum simulation of short-range spin model Hamiltonians.

A.3.5. Nanoengineered magnetic adatoms

Structures of magnetic atoms, assembled one by one on top of a surface, using the scanning tunneling microscope (STM), constitute a platform with great potential for analog quantum simulation of spin systems. Mapping this uncharted territory is the second objective of this proposal. Spin arrays can be physically separated from the conducting substrate by an atomically thin decoupling layer [HIR06, YAN17]. This reduces their interaction with the electron gas underneath, yet the resulting spin lifetime (T_1) and coherence time (T_2) are not long enough as to permit their coherent manipulation yet [BAU15, WIL18a]. Therefore, at this point, this platform can only perform quantum simulations of the equilibrium properties. As I discuss below, this is enough to explore some outstanding open problems in quantum magnetism, such as the fractional end states in spin liquids, or the simulation of ground state of the quantum Heisenberg model in a triangular lattice.

Experimental progress in the fabrication and measurement of STM-build spin-lattices has witnessed several important breakthroughs in the last few years. The potential of this approach is related to the following properties:

- **Chemical diversity.** A variety of magnetic atoms (Ti, V, Cr, Mn, Fe, Co, Ni, Gd, Ho) can be deposited in various conducting substrates, including metals, semiconductors, graphene and superconductors. In the case of metallic substrates, it is convenient to introduce an insulating decoupling layer, such as Cu₂N and MgO, to separate the magnetic atoms and the electron gas.
- **Atomic manipulation.** STM brings the capability to engineer structures, such as dimers [HIR06,YAN17], trimers[YAN17b], spin chains of variable length[HIR06,TOS16,KAM18], spin ladders[LOTH10], and even spin-chain junctions[KHA11]
- **Large scale atomic manipulation.** An obvious question is the limit for the dimensions of the structures that can be assembled using STM. So far, the largest structure made so far has more than eight thousand atoms [KAL16]. There is no physical reason that prevents going way beyond this limit.
- **Spin polarized (SP) STM** [WIE09] permits to map (relative variations of) the surface average magnetization with atomic resolution [WIE09]
- **Inelastic electron tunneling spectroscopy** (IETS) with atomic resolution [HEI04,HIR06,HIR07], permits to probe excitations in the 0.1-100 meV range, with thermally limited spectral resolution (0.1 meV). I demonstrated that IETS gives access to the surface spin spectral function [JFR09], setting the foundations for the atomic resolution imaging of spin waves in spin chains [SPI14].
- **STM pump and probe** to drive single surface spins, both individually and integrated in spin arrays. This permits to measure the spin relaxation time (T_1) of both individual surface atoms and spin arrays [LOT10,LOT12,SPI14].
- **STM spin paramagnetic resonance** (STM-ESR) [BAU15,CHO17,WIL18a,WIL18b] can probe spin excitations with spectral resolution limited by decoherence of the surface spin. The spectral resolution achieved so far is 10 neV (3MHz). As a trade-off, this technique can only probe low-energy transitions (up to 100 μ eV or 25 GHz). This resolution permits to carry out **absolute magnetometry** of adjacent atoms by doing spin resonance on a single atomic probe spin[CHO17,YAN17], as well as the splitting due to **hyperfine interactions** with nuclear spins individual magnetic atoms and dimers [WIL18b]. STM-ESR, combined with STM pump and probe, permit also to measure T_2 .

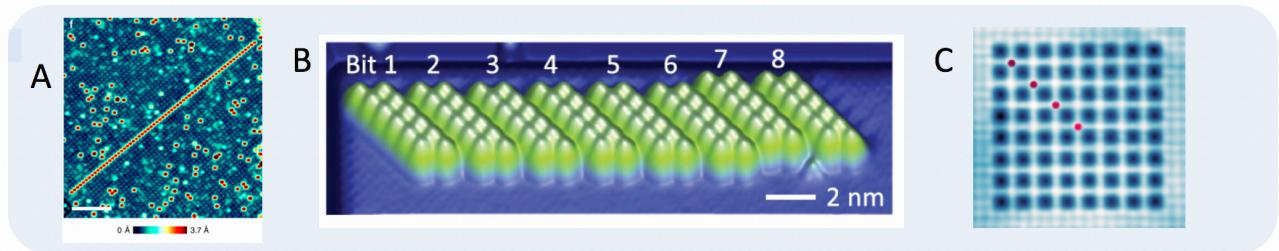


Figure 4. STM images of engineered atomic structures **A)** Chain of 60 Fe atoms on a superconducting substrate [KAM18]. **B)** Array 8 ladders with 12 Fe atoms each [LOT12]. Ladders can order in two Neel states, forming a bit. **C)** 8x8 array of Chlorine vacancies on a square lattice of Cl on Cu(100) [GIR17]

A.3.6. Analog Quantum simulation of spin models with magnetic adatoms

Standard model Hamiltonians with quantized spins provide an extremely accurate description of the experiments [HIR06,HIR07,LOT12,SPI14,YAN15,TOS16]. Thus, the spin excitation IETS of Mn spin chains deposited on Cu₂N/Cu(100) was successfully described with a Heisenberg model [HIR06] with a first neighbor exchange $J=5.9$ meV [JFR09]. Analogously, the single atom spin-flip excitation IETS of Mn, Fe o on the same surface are also very well described with single-ion anisotropy spin Hamiltonian [HIR07,JFR09,YAN15] and show that Fe has a much larger magnetic anisotropy than Mn. very different magnitude of magnetic anisotropy. Quantized spin model Hamiltonians also describe very well other surfaces, such as Fe and Ti on MgO [BAU15,YAN17].

The energy scales of the model Hamiltonians can be obtained by fitting to IETS or ESR-STM experiments, but can also be predicted from first principles calculations. From the theory side, the DFT calculations permit to build an Anderson model for the localized d orbitals, and the exact diagonalization of the model in a cluster gives many-electron states that are used to build the spin model [FER15a,FER15b]. These works show how the quantized spin of the model is host by many-body states that combine electrons in the magnetic adatom, weakly hybridized with those of the surface.

The Kondo interactions between magnetic adatoms and the substrate are an obvious source of concern when it comes to use magnetic adatoms to simulate Heisenberg-type spin Hamiltonians. When magnetic atoms

are placed directly on a metal, without a decoupling layer, Kondo effect occurs often [TER08], screening the magnetic adatom magnetic moment. This is why the decoupling layer is essential to reduce the Kondo exchange interaction. However, even if there is no Kondo effect, the remaining weak Kondo exchange still causes spin relaxation and decoherence [DEL14,DEL17] and a reshapes the IETS spectra[TER15].

Engineered Fe spin chains assembled on Cu₂N/Cu(100) STM have permitted to observe the emergence of broken symmetry FM and AF states, when they have more than 3 atoms [LOT12,SPI14,YAN17b]. The sign of the exchange is controlled by the orientation of the chain on the surface. In contrast Co chains [TOS16] and Mn chains[HIR06] on Cu₂N/Cu(100) do not show a magnetically ordered ground state. This reflects the effect of quantum fluctuations on account of the smaller magnetic anisotropy [DEL15].

The fabrication of atomic spin chain with STM permits to create chemically inhomogeneous spin chains. This includes heterogeneous dimers [CHO17], but also fabricate chains with up to 9 Mn atoms ($S=5/2$) terminated with an individual Fe ($S=2$) atom [CHO17b]. This resource permits to study the effect of magnetic impurities in otherwise homogeneous spin chains, a problem that has attracted a great deal of attention from the theory side [EGG92]. Of course it is also possible to build two different structures, made with the same atoms, close to each other [YAN17b]. In both types of composite systems, either chemical or structural composites, it is possible to readout physical properties of one of the parts of the system by performing direct physical measurements on the other. For instance, the magnetic moment of an atom can be measured by performing an ESR-STM experiment of a second atom nearby, and reading the shift of the resonance as a function of the distance to the source atom [CHO17]. The magnetic field created by a structure can also be inferred by its influence on the spin relaxation of a structure nearby [YAN17b]

The second objective of this proposal is to carry out quantum simulations of spin Hamiltonians. More specifically, I will focus on the simulation of spin liquids. They can be defined as “*a ground state in which there is no long-range magnetic order and no breaking of spatial symmetries and which is not adiabatically connected to the band (Bloch) insulator*”. The theoretical understanding of quantum liquids is a very advanced field, in particular for one dimensional systems for which efficient computational methods, such as density matrix renormalization group (DMRG) [WHI92] , and exact solution for short-range Heisenberg and Ising models. In 1D, the role of topology, the parity (half vs half integer) of the spin and their relation to a gap in the excitation spectrum the existence of hidden topological order and its connection with edge fractionalized states are settled matters by now [HAL18]. In contrast, the understanding of quantum spin liquids in 2D is much more challenging. For instance, the nature of the ground state of a triangular lattice of antiferromagnetically coupled $S=1/2$ spins is not clear yet.

The strategy adopted in this proposal is to show, from the theory stand point, how to use magnetic adatoms to carry out quantum simulations of 1D and 2D spin liquids. Probing spin liquids is very challenging. The usual experimental smoking that characterizes them are the absence of magnetic order and the absence of spin waves with sharply defined features in the (w,q) plane [TEN95,HAN12]. QUASAL proposes strategies to address this challenge. The work plan to address objective 2, has the following tasks:

Hamiltonian mapping. Here we need to find out the combination of magnetic atom and decoupling layer that produces a given quantized spin S , the desired short range antiferromagnetic exchange interactions, that permit to fabricate artificial lattices with the desired geometry (chains, ladders, square arrays, triangular arrays). This task is accomplished using a combination of DFT calculations, derivation of model parameters and exact numerical diagonalizations [FER15b]. MgO and hexagonal boron nitride are optimal decoupling layers to obtain square and triangular lattices respectively. Another aspect to be worked out is the mitigation of the Kondo exchange with substrate electrons. This can be accomplished with thicker decoupling layers as well as using superconducting substrates [HEI13].

Hamiltonian control. The main control parameters are: 1) the structural design of the lattice , 2) the magnetic field, 3) the exchange field of the tip[LAD17b,YAN17] 4) the electric field of the tip [BAU15]. Structural control includes the lattice geometry (square, triangular, honeycomb) and the dimensionality (chain, ladder, N chains, 2D array) and the number of atoms.

Detection. An essential part of quantum simulation entails probing the simulating platform to infer information about the simulated system. Nanoengineered spin chains offer a good opportunity to simulate their spin correlation functions of $\langle S_z(0) S_z(j) \rangle$. For that matter, I will explore a detection strategy, shown in **Fig. 5**, that involves the extensive use of structures with additional atoms (red balls) inserted as magnetic impurities with very large magnetic anisotropy, , to act as sources of a local magnetic field, or nearby, to act as magnetic sensors [CHO17]. The inside impurity induce a finite average local magnetization that can be

probed either with a spin polarized tip [LOT12] or by performing an ESR-STM experiment on an external magnetic atom [CHO17]. This approach provides a direct measurement of the static correlation functions. Note that both the impurity atom as well as the external probe atom can be placed in different locations, providing additional information. In addition, dynamical spin fluctuations can be picked up by their influence in the relaxation and decoherence time of a nearby spin [YAN17b, DEL17, ITO08].

Validation. In the case of chains and ladders, DMRG can be used efficiently to compute correlation functions[WHI92], and spectral functions[HOL11]. Therefore, accurate predictions for a given system can be done, combining model building and DMRG. The problem of the ground state phase for the Heisenberg model on triangular 2D lattice will be tackled using exact diagonalization on a small cluster and using DMRG in cylinders (stripes with periodic boundary conditions).

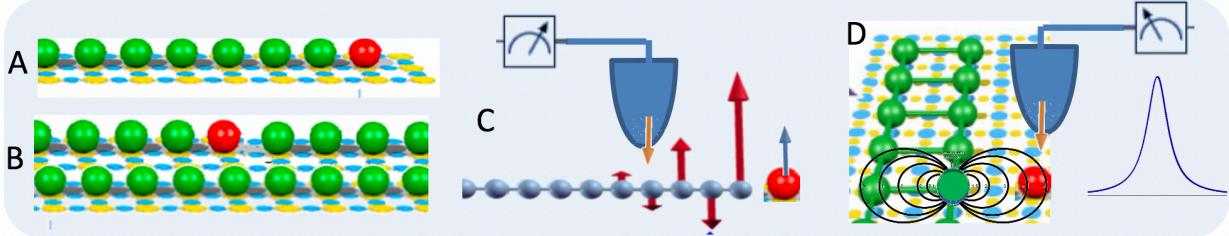


Figure 5. Quantum simulation of spin liquids. (A) and (B): A chain and a ladder, made with $S=1/2$ atom (Ti on MgO), [YAN17], with a magnetic impurity, red ball, with very large magnetic anisotropy (for instance Co [RAU14]) in the edge(A) or in bulk(B), that induces a local magnetization. (C) SP-STM probing magnetization in A. (D) Nearby probe atom (red), such as Fe is used to probe spin noise and/or magnetic field created by spin ladder. Figures adapted from [CHO17b] & [YAN17b]

A.3.7. Optically induced exchange interactions: state of the art.

Objective 1&2 employ an equilibrium approach. This is forced by the lack of a fast mechanism to switch on and off exchange interactions in a time scale shorter than the decoherence time of the simulation degrees of freedom in the proposed platforms. Whereas the equilibrium approach can be enough to probe equilibrium of properties of quantum matter, there quest for exchange interactions that can be turned on and off is an obvious next step in the roadmap for quantum simulation, both analog and digital.

Objective 3 looks for a viable solution for accurate time control of exchange interactions between quantum spins. This would be for instance a resource to implement two qubit gates in the case of exchange coupled $S=1/2$, a prerequisite for universal quantum computations. In the case of spins localized in semiconductors quantum dots, most of the experimental and theoretical work has focused on the electrostatic modulation of the tunnel barriers separating two dots [LOS98]. However, this approach has been notoriously difficult and the largest number of dots controlled in this manner is only nine [MOR18]. Therefore, the quest of alternative strategies is needed.

In 2002 the group of professor Lu Sham proposed a new mechanism to induce exchange interactions between distant localized spins in semiconductors[PIER02]. They showed that a laser with frequency below the band gap induces an exchange coupling between local spins, mediated by the population of virtual excitons[PIE02]. This so called optical RKKY (ORKKY) is proportional to the Rabi coupling with the laser, the inverse of the detuning frequency, the excitonic strength and the exciton-local moment exchange. The theory was originally proposed[PIE02] to control the spins of electrons localized in quantum dots. I did extend the theory to the case of magnetic atoms in semiconductors [FER04], and to the case of ORKKY mediated by microcavity polaritons[QUI06], that permits to extend the range of the useful interactions up to half a micron.

Indirect experimental evidence of ORKKY interaction has been reported for an ensemble of InGaAs quantum dots[SPA11]. The reported magnitude of the laser induced interaction was $J = 1 \mu\text{eV}$. If ORKKY interaction is to be used in analogue quantum simulations, this implies that very low temperatures have to be used, otherwise $kT > J$, and the system will always be in the entropy dominated phase. Thus, increasing J is a strategic goal to make practical use of ORKKY.

A.3.8. Optically induced exchange interactions: a new approach

The disappointing experimental status of ORKKY, together with its large added value as a resource for quantum simulations, call for a radically different approach. I claim that the key issue here is the choice of the material system to enhance the magnitude of ORKKY. Therefore, I propose to explore the ORKKY in two dimensional transition metal dichalcogenides (TMD), such as MoS₂, WS₂, MoSe₂ and WSe₂. These are

semiconducting crystals, with band-gaps in the range of 1.5 to 2 eV, the same energy range than II-VI and III-V optoelectronic materials of choice (GaAs, CdTe), but with physical properties that make them ideal suitable to implement ORKKY:

- 1) **Very large excitonic effects**, that enhances the optical response, and thereby ORKKY [PIE2]. Exciton binding energy is much larger in 2D transition metal dichalcogenides than in conventional II-VI and III-V semiconductor quantum wells, with exciton binding energies are in the hundreds of meV, compared to 10 meV for GaAs quantum wells.
- 2) **Large intrinsic light-matter Rabi coupling**. In transition metal dichalcogenides this can easily exceed by a factor of 2 that of conventional GaAs and CdTe semiconductors. Thus, the reported polariton splitting for MoS₂ microcavities reaches 46 meV [LIU15], whereas the same quantity in GaAs and CdTe microcavities is in the range of 15-25 meV [SAB01].
- 3) **The two dimensional nature** is a huge advantage to minimize unwanted optical absorption of sub-gap radiation, compared to quantum wells embedded in bulk systems. This permits to use larger laser power.
- 4) **Functionalization of 2D crystal**, by adsorption of molecules, nanoislands, and so on, permits an additional route to provide local spins, in addition to doping. In particular, I will explore the coupling between spins known to have a extremely long spin coherence times, such as endohedral N@C₆₀ complexes with record T₂ times [MOR07] (see figure 1C of this proposal).

The work plan to address objective 3, has to address the following specific objectives:

Hamiltonian mapping. For a given 2D crystal and a given localized spin, we need to compute the ORKKY Hamiltonian, that can have both symmetric, non-symmetric and antisymmetric exchange contributions, and an effective Zeeman field, depending as well on the light polarization. A great deal of work will have to focus on the modelling of adsorbates and point defects. The target is to look for large T₂ adsorbates that have a mild interaction with the 2D crystal, so that both spin and optical properties of the two parties are not radically modified, yet their coupling is not negligible.

Hamiltonian control. The ORKKY depends on some parameters that can be easily changed, such as laser power and frequency, and also on the specifics of the system. For instance, we demonstrated that ORKKY is very different if the 2D crystal is embedded in a cavity, so that the mediating particles are optical polaritons [QUI06]. Another source of control could come from the in-plane modulation of the ORKKY intensity arising in twisted bilayers of 2D semiconductors that lead to a moire pattern that modulates the band-gap in-plane. The spin decoherence time of the adsorbates, due to coupling to the phonons, thermally excited carriers and nuclear spin of the host will be computed [DEL17] to assess if coherent optical control is possible.

Detection of ORKKY. The approach here is to explore the variations of conventional electron spin resonance (ESR) experiments in response to the sub-gap radiation. As the ORKKY interaction is ramped up, it should induce either a motional narrowing or a broadening of the ESR line. This will be modelled carefully for the specific platforms where ORKKY interaction is predicted to be sufficiently large.

Section b. Methodology and Work Plan

This proposal has a unique grand goal: to pave the way to build analog quantum-simulation solutions to the quantum many-body problem. For that matter, the proposal defines, from the theoretical point of view, the roadmap to carry out analog quantum simulations using 3 different physical platforms. The work plan addresses: 1) the design of the simulating systems, 2) the modelling of experimental probes that are feasible with state of the art and how they convey information of the simulated system, and 3) the validation of the simulated results.

The workplan requires a variety of methods I have used over the years, such as density functional theory (DFT) based calculations of the electronic structure [JFR05,KOS13, FER15, CAR18b ,SOR18], post-processing techniques to obtain tight-binding descriptions [KOS14,FER15, FER15b,LAD16, LAD17, SOR18], exact diagonalization of models [JFR06, JFR09,DEL13, FER15a, FER15b], large scale quantum transport calculations [MUÑ09,LAD13], STM transport and IETS [JFR09, DEL11, DEL13, KLE18], STM-ESR modelling [LAD17b,WIL18a,WIL18b], open quantum system decoherence calculations [DEL12,DEL17, DEL17b], optical response calculations [JFR06,LEG06,BES12], optical RKKY calculations [JFR04,QUIN06] . I will also use some recent additions to my toolbox, such as **machine learning** [CAR18] and Density Matrix Renormalization Group [BAN17]. In addition, I will use some very

novel techniques, such as **digital quantum simulations on real quantum computers**, that we are starting to explore in my group [GAU18].

Importantly, I have very extensive experience and very cited contributions in the three systems of interest (graphene [JFR07], magnetic adatoms[JFR09] , dichalcogenides[KOS13,KOS13b]). I did seminal work on the ORKKY interactions [JFR04] and on the potential for applications of hydrogenated graphene[SOR10]. In 2015 I organized a SPICE workshop on *Magnetic Adatoms as Building Blocks for Quantum Magnetism* [<https://www.spice.uni-mainz.de/maqm-workshop-2015/>] that brought together experts on these two different communities (surface science and quantum magnetism), that has provided a well documented background for this proposal.

B. 1 Methods for Hamiltonian Mapping.

Hamiltonian Mapping consist on 1) the demonstration that the low energy states of a given physical system are described by a certain model Hamiltonian (Hubbard, Heisenberg), and 2) the calculation of the energy scales of the model. The systems are defined by a host material (graphene bilayer, MgO bilayers on silver (100), transition metal dichalcogenide) interacting with some point defect (sp^3 functionalization, magnetic adatom, molecular adsorbate) (See Figure 1).

B1. 1 Hamiltonian mapping for the Hubbard model

The derivation of the Hubbard model parameters associated to a functionalized graphene bilayer is done as follows:

A **tight-binding model** for the unit cell containing either a single point defects is generated (see below for more details). The size of the unit cell has to be much larger than the dimension of the bound state, which can easily reach 10nm. Therefore, the number of atoms in the unit cell can be as large as 98.000 A **Lanczos numerical diagonalization** is carried out to obtain the single-particle states close to the Dirac energy. This permit to obtain the gap induced by the electric field, the energy of the in-gap state, and its wave function.

Computation of Hubbard U , as the Hartree integral associated to the in-gap orbital.

Computation of hopping t . This can be done repeating the procedure for a unit cell with two defect sites. This yields a couple of in-gap states, that are the bonding-antibonding states associated to the hybridize in-gap states. As I showed recently [ORT18] a canonical transformation permits to obtain the single defect states, as well as the hopping integral between them.

Computation of other many-body terms. The magnitude of additional many-body terms in the Hamiltonian [JAN14], such as the inter-site exchange, correlated pair hopping and density assisted hopping, can be done representing the Coulomb operator in the basis of the two defect states.

Computation of energy bands. As a complementary strategy to point 4, we can use periodic boundary conditions to obtain the energy bands that describes an ordered lattice of point defects. The results of this procedure are shown in figure 3C for “artificial graphene”. Notice that the bandwidth of the artificial graphene is just 15 meV in the preliminary results, and it could be made smaller by increasing the lattice spacing, preserving the same value of U . The hopping matrix for the artificial lattice can be obtained by numerical fitting to a model with 1 orbital per impurity site and hopping up to third neighbors . The results of the fitting are also shown in figure 3.

Regarding step 1, a full DFT calculation might be out of reach, due to the large size of the crystal, but it is not necessary. In the case of hydrogenated graphene, there is a simpler strategy. A few years ago we carried out DFT calculations for hydrogenated graphene and showed that they give results very similar to those of a 1-orbital tight-binding model where the chemisorbed hydrogen is modelled as a missing site in the lattice[SOR10]. Part of the work plan includes the study of sp^3 functionalization using planar molecules that can self-assemble to form an ordered array, going around the need to position the sp^3 centers one by one. For this task, it will be necessary to carry out DFT calculations with a very large number of atoms.

B1. 2 Hamiltonian mapping of spin models

The choice of magnetic atoms and surface, define the class of quantum Hamiltonian that can be simulated. The procedure to predict the spin Hamiltonian that is simulated by a given choice of magnetic atoms and surface is based on our recent work, showing a reliable method [FER15,FER16b] to derive spin model Hamiltonians starting from a DFT based calculation of a point defect/molecular adsorbate embedded in an otherwise pristine solid. The method has 3 well defined steps (See Figure 6) :

DFT calculations in order to have accurate atomic positions and chemically correct description of the electronic structure of the system. This entails DFT based calculations of the system, with a large unit cell

that contains one or several defects. I have ample expertise in this type of calculations [FER15b, FER16, WIL18]. DFT methodologies are based plane wave and linear augmented plane waves basis sets are more reliable than atomic basis set. This makes it necessary to implement the next step.

Wannierization in order to represent the Kohn-Sham Hamiltonian in a basis set of localized orbitals. The Hamiltonian of the point defect/adsorbate/magnetic adatom becomes a multi-orbital Anderson model[FER16b]. This is accomplished using the exact mapping between Bloch states of the DFT Hamiltonian and the basis of maximally localized Wannier orbitals. When this procedure is implemented successfully, the Wannier basis describes a set of atomic orbitals centered around the atoms (see for instance my work on transition metal dichalcogenides [KOS13]).

Mapping to a spin model. The Anderson model can be mapped into a spin model using two approaches. In the case of magnetic atoms/adsorbates in insulating hosts, it is possible to truncate the many-electron configuration space and carry out an exact diagonalization in the restricted space. The resulting wave functions permit to build the parent spin model [FER15, FER16]. For conducting hosts, a Schrieffer-Wolff transformation yields a Kondo model.

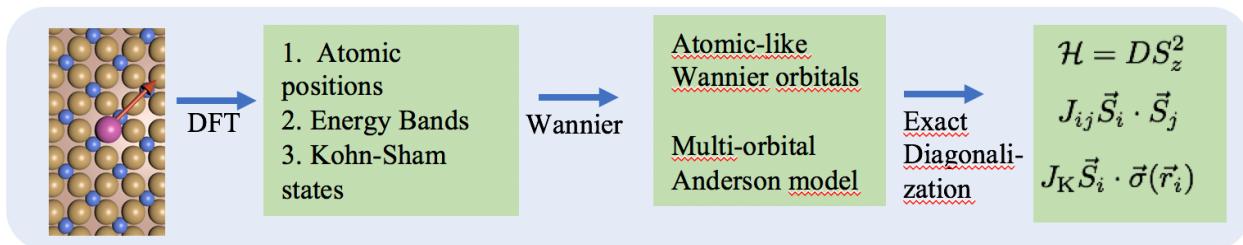


FIGURE 6. Spin Hamiltonian Mapping flow chart.

We have extensively used this approach to obtain an accurate prediction of the single ion anisotropy [FER15a, FER15b, LAD17b] as well the Kondo exchange with the delocalized states [FER15b]. When applied to a unit cell with two impurities, this can also permit to compute exchange between the two local spins. We have also obtained exchange directly from DFT, computing ground states with different magnetic ground states (see for instance my work [LAD17, SOR18]).

I will consider mostly surfaces that have been already tested experimentally: Cu₂N/Cu(100) [HIR06, HIR07, LOT12, SPIN15, CHO17b, YAN17b], MgO/Ag(100) [RAU14, BAU15, PAU17, YAN17, WIL18, WIL18b], and h-BN/Ru [JAC15]. This choice still leaves a huge exploration space in terms of magnetic atoms to deposit. I target systems with S=1/2 and S=1. For S=1/2 this can be accomplished in two ways, either with systems with a real S=1/2, (such as Ti and Cu on MgO [YAN17, YAN18] or some low energy doublet from a higher spin system, such as Co on Cu₂N [TOS14]. For S=1 a goal of this project is to find systems with a sufficiently low magnetic anisotropy to test predictions, such as the fractional end-states in Heisenberg spin chains[DEL13].

B1.3 Hamiltonian mapping for the Optical RKKY

ORKKY interactions are mediated by virtual excitons: in the presence of sub-gap laser radiation, the ground state of the solid gets dressed with states with a finite density of excitons. The amplitude of the dressing depends on the intensity of the laser, and it does not entail any absorption of energy. The starting point for the calculation of the ORKKY is a Hamiltonian that describes the conduction and valence bands of the semiconductor, their exchange coupling to the local spins, their coupling to the photon field, and the electron-hole interaction [FER04]. Therefore, the first step is to derive such a Hamiltonian, using the methods outlined in B1.1. The ORKKY interaction is calculated by integrating out the fermionic degrees of freedom, dressed by their coupling to the radiation, and including the electron-hole excitonic effect, that enhances strongly the ORKKY [PIE02].

Thus, a predictive calculation of the ORKKY has the following steps:

DFT calculation + Wannierization+ Schrieffer-Wolf (see Figure 6 and sec. B.1.1) to obtain a Kondo model for the local spins coupled to the conduction and valence states of the host semiconductor. Here is where the role of the specific host semiconductor and spin-dopant are accounted for.

Solution of the excitonic problem, using exact diagonalization[JFR06] in the sub-space of one electron hole pair that occupy the states obtained in the previous step. This yields a material dependent the renormalized Rabi coupling [JFR04]

Computation of exciton-polariton basis (for cavities) or dressed exciton basis (otherwise). In both cases (cavities, else) this can be done treating photons fully quantum mechanically, as we did in our work, which defines a coupled-boson problem that can be diagonalized exactly. In the case exciton-mediated ORKKY it is also possible to treat the laser light classically [JFR04].

Transformation the Kondo coupling to the polariton/dressed exciton basis. This change of basis (see my work [QUI06]) yields a Hamiltonian for local spins interacting with polaritons/dressed excitons.

Integrating out the polaritons/dressed excitons. This yields a spin-only Hamiltonian with the ORKKY coupling as a function of material parameters, local spins, laser power, polarization and frequency. This last step can be carried out diagrammatically [PIE02], semiclassically [JFR06] or using canonical transformations[QUI06]. Here I will do it using the Bloch-Redfield formalism[DEL17b] for open quantum systems (see for instance eq. 17 &18 of my recent work deriving conventional RKKY in this manner [DEL17b]). This will permit to compute both the ORKKY as well as the spin relaxation and decoherence induced by this coupling [DEL17b]

B. 2 Modelling for Hamiltonian control, probing and validation

Once a simulating platform is build, the next stage involves 3 steps. First, the control of some physical magnitude that translates into a change in an energy scale of the simulated Hamiltonian. Second, probing the system and relating the response function in the simulating platform to properties of the simulated Hamiltonian. Third, a comparison of the simulation performed with some alternative method to model the Hamiltonian. In general, the latter is only available either in 1D systems or in finite size systems.

B2. 1 Hubbard simulating platform: modelling control, probing and validation

The best control parameters in the functionalized graphene bilayer are the position of the sp^3 defects, that define the lattice to be modelled, as well as the strength of the electric field, that modulates the extension and sublattice polarization of the in-gap states, and thereby their hopping and t and Hubbard U . The method described in B1.1 applies here as well.

Transport is the most obvious probe for functionalized graphene bilayer. I will simulate both STM transport, that permits to obtain the spectral function of the bilayer, and in-plane transport. STM transport can provide a map of the density of states, and if this one vanishes at low bias, as in the insulating phases of the Hubbard model, cotunneling will provide the spectral function of the electronic collective modes [DEL11]. In plane 4-terminal transport can provide information about superconductivity, that could arise in the square lattice, slightly away from Half filling [AND97], and Hall effect, that could arise in the triangular lattice at $\frac{3}{4}$ filling [MAR08]. For reference purposes, in-plane transport simulations will be carried out first ignoring interactions, using Landauer formalism and advanced methods to solve the Dyson equation [LAD13]. In order to include the effect of interactions both on the STM and in-plane transport , I will carry out a brute force numerical diagonalization, possible only in the case of small simulation arrays. In the case of 1D structures, I will use DMRG to compute linear conductance [BIS17].

The validation of the simulations will be carried out by testing small clusters of sp^3 centers for which an exact diagonalization of the model is possible. For instance, a transport experiment can yield an observed transport gap that is tested against the predictions of theory. This permits to calibrate the simulating platform. Once we are sure this works for small systems, we can repeat it for larger scale systems, still within the range of exact calculations. This should confirm that up-scaling the dimension does not compromise the simulations. Once this is done, the experimental platform can be used to simulate systems out of reach of conventional numerical calculations.

B2. 2 Spin liquid simulating platform: modelling control, probing and validation

For a given choice of magnetic atoms and surface, the control parameters are: 1) geometry (chain, ladder, array) of the structure, 2) the external magnetic field, and 3) the influence of the tip on the structure, in the case of magnetically functionalized tips (see for instance my papers [YAN17,LAD17b]). The methods for modelling this are the same than B1.2.

The strategy for probing spin liquids has to circumvent their notorious lack of clear experimental smoking guns. Here I will target the simulation of the static spin correlation function and the excitation spectra. For the correlation function I am proposing a new strategy, only possible thanks to the recent developments in atomic manipulation and ultra-precise ESR-STM. The strategy, depicted in Figure 5, is to introduce a local magnetic atom with large spin and magnetic anisotropy that, induces a spin polarization, via exchange on one atom of the spin liquid. The magnetization, that might be very small, creates a magnetic field that is

now peaked by a probe atom that is placed nearby the target structure. The probe atom can be moved to several locations, to map the magnetic field created by the structure.

In order to assess this strategy to simulate quantum spin liquids, the tasks for the theory are: 1) to compute the magnetization induced in the spin array. 2) To compute the magnetic field that this creates 3) To determine in how many sites it is necessary to carry out the ESR-STM measurement. Task 2, will be done by exact diagonalization in small size systems and using DMRG in the 1D case (chains, ladders). An example of this type of calculation can be found in my paper [BAN17]. Task 3 is essentially a problem of image recognition: for a given input (the magnetic field measured in a set of points) we'd like to determine the distribution of source magnetic moments. I will use supervised learning with neural networks for this task. [CAR18].

An unperturbed spin liquid has zero magnetization with a characteristic noise spectrum, associated to its excitation spectrum. The noise spectrum can also be probed by its effect on the T_1 and T_2 of a nearby probe atom. The principle is the same than the one used when nuclear T_1 is measured as a probe for spin dynamics of the spin liquid [KUR05]. The calculation of T_1 and T_2 will be carried out using the same Bloch-Redfield methods we used in [DEL17]. We shall also explore if the spatial modulation of IETS spectra recorded in different atoms in a $S=1/2$ spin chain can provide evidence of the spinon quasiparticles by looking at the formation of spinon standing waves, in analogy with my work for spin waves in spin chains [SPI14].

For the validation of the simulations, the extensive literature on 1D systems will provide a very good benchmark to test the predictions of theory. In addition, I will explore the use of Digital Quantum Computations, in the quantum computers that are available online (IBM, Rigetti) to model spin model Hamiltonians, using the quantum phase estimation and the quantum Fourier transform algorithms [SAL15]. At the time of writing this proposal we are doing our first efforts along this line in my group [GAU18].

B2. 3 Optical RKKY platform: modelling control and probing and validation

Control of ORKKY interaction, in a given system, is given by the laser frequency, power and polarization state. This will be modelled using the methods described in B1.3. An important task for theory, to be addressed here, is to propose methods to detect that ORKKY interaction is taking place. This should be done first in samples where no attempt to create an ordered array of local spins is done. The detection strategy will be based on spin resonance. For that matter, I will study the effect of the ORKKY on the microwave absorption spectra of the system. The detection of that experiment could be done in a conventional setup, for very large area samples, or using spin-noise Faraday spectroscopy [CRO04]. A task for this project will be to develop a theory for this type of spectroscopy in spin-doped transition metal dichalcogenides.

B3. Summary of work plan

In the following table I list the tasks, methods and expected timing for this proposal. More info in the text above.

	OBJ 1. Hubbard.	Year	OBJ2. Heisenberg	Year	OBJ3. ORKKY	Year
Hamiltonian Mapping	Tight Binding. Lanczos	1-3	DFT+ Wannier: 	1-5	DFT+ Wannier:	1-5
	Calc. U, t, other	1-3			Exciton by Diag.	1-3
	DFT+ Wan.:	3-5			Dressed basis	3-5
Control	Tight. Binding.	1-3	Tip exchange DFT	3-5	All of the above	3-5
Modelling detection	Landauer transport:	2-3	DMRG Correl. Func.	2-3	Normal ESR	1-3
			DMRG spin noise.	3-5	Spin noise spectroscopy	3-5
	DMRG transport	3-5	Machine Learning	4-5		
Validation	DMRG, & Diag	3-5	DMRG	1-3	Not. Applicable	
	Digital Quant. Sim.	1-5	Digital Quant. Sim	1-5		
Host, defect	Graphene bilayer, sp ³ defects		Cu ₂ N, MgO, h-BN, Ti,Cu,Fe...		MoS ₂ , WSe ₂ ..etc, N@C ₆₀ ..	

Section c. Resources (including project costs)

The completion of this proposal will rely on the formation of a very strong team of researchers, guided by the PI, that will be supported by a computer system administrator. The team will be organized in 3 groups, dedicated to each one of the 3 objectives. The composition of the groups will have 1 PhD student and 1 postdoc (INL fellow). The reference duration of contracts of all the postdocs and PhD students will be 3 years. There will be 2 generations of teams, starting on years 1 and 3. Therefore, on year 3 there will be an overlap between the researchers of the first and second half of the project, that will ensure the transfer of know how between the two generations. This breakdown is implicit in task timing in the table above.

I am requesting funds to cover 50% of my salary, matching my dedication to the project. I am also requesting the 50% of salary of a system administrator to provide assistant on the management of the computer cluster, software installation, software debugging.

In terms of equipment, the project makes intensive use of computational work. INL counts with a computational cluster, but I am requesting 60k€ to upgrade in order to avoid oversubscription. In addition, we will make use of external supercomputers, provided by the Spanish Network of Supercomputers (*Red Española de Supercomputación*). This requires to write a proposal that, in my case, have been always approved, granting free access to computational resources that are adequate for the numerical work required by QUASAL.

I am also requesting travel budget on a basis of 3 travel per researcher and year, at an average rate of 1.5k€ per trip (157.5k€). In addition, I am also including 40k€ to invite researchers to visit the group, engage in collaborations, and also to organize an international workshop on “Quantum Simulation in Artificial Lattices” at INL in the third year of the project, to disseminate results and gather feedback from the community (estimated cost 20k€).

The budget for dissemination will be used for publication costs (40 publications at an average of 1.5k€), the creation of a specific website for QUASAL (10k€) and the subcontracting of 2 two outreach videos on quantum simulations and artificial lattices (10k€ in total).

The last, but not the least, my group at INL counts with the standard necessary resources, such as office space and material, books, meeting rooms, access to on-line scientific journals, administrative support, in the great facilities of INL campus, that include a residence for visitors and a creche for the children of our employees. INL counts with a staff to take care of the project management, financial reporting, human resources support. The most important of all, INL offers a unique scientific atmosphere, that fosters high quality research and brings together researchers from all over the world to work in the only international organization dedicated to do research in Nanoscience.

Budget Table

Direct Costs				Indirect Costs		
Personal	k€	Other	k€		k€	Total ELIGIBLE
PI	210.6	Travel	153.5	B. Overhead	498.1	
Postdocs	756.0	Equipment	60			2.498.459,0€
PhD	561.6	Publication	60	C1 Subcontracting (No overheads)	8000	TOTAL REQUESTED
Admin+ System Man.	131.2	Other (Web, Videos)	60			
TOTAL Personal	1659.4	TOTAL Other	333.5	C2 Other Direct (No overheads)		2.498.459,0 €
A. TOTAL DIRECT						
1997,5						

Please indicate the duration of the project in months:	60
% of working time the PI dedicates to the project over the period of the grant:	50 %

REFERENCE LIST (B2)

(It does not count towards the page limit)

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