Programme de recherche

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Je présente dans ce document mes perspectives de recherche pour les années à venir, regroupées en deux directions différentes.

- La première partie (Partie I) concerne la réponse non-lineare à des champs multiples et forts. Je souhaite m'attaquer frontalement a le problème de **la génération d'harmoniques d'ordre élevé(HHG)** et de **high-order sideband generation** dans les solides. J'ai la possibilité de simuler des phénomènes non perturbatifs à partir de principes primes comme HHG et d'étudier comment les effets de corrélation les affectent. De plus la possibilitè d'inclure plus d'un champ externe dans la simulation, cela me permettra d'étudier phénomènes d'interfèrence comme le HSG. Ce type d'études un laser crée une quasi-particule excitée puis un second laser les fait entrer en collision et l'émission de lumière est mesurée. Enfin, il y a une partie purement thèorique sur **l'extension de la fonction de Green hors équilibre dans le régime des champs forts**.
- La deuxieme direction (Partie II) concerne l'utilisation d'impulsions laser ultra-courtes pour contrôler l'aimantation. La technologie laser s'est développée de façon explosive jusqu'à atteindre des durées de laser de quelques femtosecondes (fs), conduisant à une nouvelle ère de physique de la matière condensée hors équilibre. **Le contrôle de l'aimantation au moyen de lasers ultra-courts** est l'une des applications les plus intrigantes dans ce domaine. En partie il s'agit d'un changement thèmatique pour moi, ayant demandé un investissement et une prise de risque importants, notamment dans un sujet très concurrentiel.

Contents

1. Solids under strong laser fields

In the last years the European Community invested a large amount of money in the construction of the X-ray Free-Electron-Laser in Hamburg (XFEL). According to their view the XFEL will open up areas of research that were previously inaccessible. Using the X-ray flashes of the European XFEL, scientists will be able to map the atomic details of viruses, decipher the molecular composition of cells, take three-dimensional images of the nanoworld, film chemical reactions, and study processes such as those occurring deep inside planets. The construction of a such large facility has been motivated by the breathtaking advances in the field of ultra-fast phenomena of the last decades thanks to the progresses in laser technology. In fact the laser duration has been constantly decreased, recently reaching the attosecond domain[1]. At the same time, the concentration of light in space and time can reach very high power-flux densities, where combined non-linear and non-adiabatic phenomena occur. The key aspect of such phenomena is the possibility of observing the real-time electronic and atomic dynamics following the excitation by means of a strong and ultra-fast laser pulse. In practice, this dynamics is connected to the relaxation of an highly excited state through several decay channels like, e.g., multi-electron-hole excitations, collective modes, phonon excitations, charge transfer, and even formation or breaking of chemical bonds and molecular rearrangements. These microscopic mechanisms, which are known to take place on extremely short timescales (from 10 to 1000 fs, see e.g. ref. [2, 3], are the basis for numerous potential applications, from solar energy to nanotechnology, biochemistry and life science. Despite the great scientific interest and the relevant technological applications, the interpretation of several key aspects of the ultra-fast dynamics following the primary excitation pulse is still difficult. Indeed, the dynamics of the initial ensemble of electron-hole pairs excited by the laser pulse represents a theoretical and numerical challenge, whose description in realistic materials is well beyond the capabilities of many state-of-the-art first principles approaches, and up to now it has been mostly based on ad-hoc models and/or approximations. Finally, by reaching the attosecond timescale, experiments can approach a new temporal regime where the dressing-up of correlations is not instantaneous, thus making the gap with theory even more profound. From these premises, it is clear that the possibility to study non-perturbative phenomena from first-principles will be of great interest for the scientific community.
In the first part of my research program I will address one important phenomena that emerged in recent years thanks to the strong laser pulses: the high-harmonic generation(HHG) in solids. I will present different extensions of the formalism, I developed in the last years to study non-linear spectroscopy, to simulate non-perturbative phenomena in bulk materials induced by strong-laser pulses. These advances will be applied to the study of HHG and other phenomena induced by light in bulk materials.

1.1 Non-perturbative response and high-harmonic generation (HHG)

During the last years I developed a new framework based on real-time solution of an effective Schrödinger equations to study non-linear response in bulk materials. In this framework the coupling between electron and external fields is described by means of the dynamical Berry's

phase and correlation effects are derived from non-equilibrium Green's functions. This approach allowed me to study second and third harmonic generation in solids and nano-structures. All these advances have been implemented in a free-available computational code: Lumen [`http://attaccalite.com/lumen`].
Until now I used only a fraction of the capabilities of this computational framework. In fact the real-time dynamics is not perturbative in the external field and in principle it can handle multiple fields with different shape, length and intensities. In order to study lowest order response functions we mainly employed sinusoidal fields in such a way to filter out the response functions at the frequency we are interested in. In a way similar to what it is done in the non-linear optics experiments. Now I would like to generalize this approach to deal with strong external fields in such a way to study new non-linear phenomena as high-harmonic generation (HHG).
HHG in isolated atoms and molecules has been the foundation of attosecond pulse metrology, extreme ultraviolet (XUV) photonics, and molecular orbital tomography. The observation of non-perturbative high harmonics generation from strongly driven bulk crystals has motivated new research aiming to probe the electronic structure of solids including normally unoccupied conduction bands, and to overcome the drawbacks of conventional XUV sources. In fact gas phase XUV sources suffer from low efficiency and therefore do not provide sufficient flux desired for many applications such as metrology and imaging. Solid-state HHG has the potential for high efficiency and high stability because of the use of high-density and rigid target as interaction medium.
Since the original discovery in single-crystal ZnO[4] several crystalline solids have been used for HHG. The important findings, such as high-energy cutoff scaling with the field[4], emergence of a secondary plateau,[5] and novel ellipticity dependence,[6] indicate that the underlying electron dynamics are markedly different from the three-step recollision model, which is widely accepted for atomic and molecular HHG. These fundamental differences are attributed to the high density and periodicity present in bulk crystals as the field-driven electron is always in the proximity of the Coulomb potential. In order to incorporate the fundamental solid-state response, two major mechanisms have been considered for HHG: the emission from nonlinear inter-band polarization and the intra-band current. Nowadays there is still an intense debate on which is the most important mechanisms for HHG, and a clear answer did not emerge yet. Beyond this topic, many other questions are still without an answer in HHG: the role of defects in HHG; how to optimize HHG in solids; effect of reduced dimensionality on HHG; HHG from amorphous solids; which information can we extract from HHG; etc..

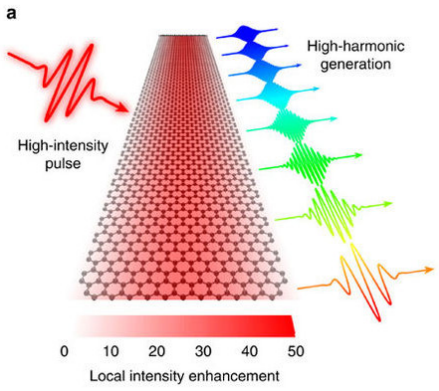

Figure 1.1: Schematic representation of high-harmonic generation in a bulk material.

Theoretical studies of high-harmonic generation are not easy. In fact due to the strong laser pulses employed in this kind of experiments, multi-bands transitions are present and low energy models are difficult to apply. Real-time simulations based on time-dependent density functional theory (TD-DFT)[7] are emerging as possible route to attack this problem. However TD-DFT have difficulties to describe correlation effects in the optical response of solids.
For all these reasons I would like to apply the formalism I developed for non-linear optics to HHG. I will have the possibility to simulate HHG response in different materials electron-hole interaction and quasi-particle renormalization.
I expect that a bunch of new physical phenomena are ready to by discovered.

1.2 High-order sideband generation

In solids, many-body correlations lead to characteristic resonances called quasi-particles, such as excitons, polarons and Cooper pairs. The structure and dynamics of quasi-particles are important because they define macroscopic phenomena such as Mott insulating states, spontaneous spin- and charge-order, and high-temperature superconductivity. However, the extremely short lifetimes of these entities make the study of their properties and interaction quite cumbersome. Recently in order to explore ultrafast quasi particle interaction and collisions, scientists exploit lightwave-driven charge transport in the time domain: a femtosecond optical pulse creates excitonic electron-hole pairs while a strong terahertz field accelerates and collides the electrons with the holes. The underlying dynamics of the wave packets, including collision, pair annihilation, quantum interference and dephasing, are detected as light emission in high-order spectral side-bands of the optical excitation.[8] A full quantum theory of these phenomena remains a challenge. Simple models in one-dimension have been proposed, but the full dynamics of electrons in a solids has never been simulated. This requires advanced capabilities to follow electron-hole dynamics in real-time including correlation effects that are responsible for the present of quasi-particles. Moreover in the experiments[8] it was argued that a correct description of dephasing effects on short time-space is a necessary ingredient to explain and address these phenomena. At the beginning I plain to use a phenomenological dephasing. Later, by means of the merging of Berry-phase formulation a Green's functions (see next section), dephasing effects will be derived in a rigorous way.

1.3 Merging Berry-phase with Non-equilibrium Green's functions

The last topic of this first part of my research program is a pure theoretical work. The problem I would like to solve is the merging of Berry's phase theory and non-equilibrium Green's functions. The problem is easy to describe. On one hand the coupling between electrons and external field in extended systems is described by means of Berry phase. On the other hand many-body perturbation theory based on Green function allows to describe correlation effects in bulk materials in a very efficient way. The problem is that combination of these two world is not an easy task. If fact the Berry's phase is evaluated by the change of wave-function phase, while in the Green's function formalism all information about the wave-function phase is lost.[1] Different solutions have been proposed in the literature, passing through the current,[9] increasing the dimension of space[10], but none has prevailed over the others. My plain is to test the different approaches proposed in the literature and see if it is possible to improve or simplify them, or to propose a new one. The successful realization of this project will allow to include dephasing effects derived by NEGF in the strong field regime. Moreover the results will be important also for the second part of this research program. In fact, the magnetization that is at the core of the second part of the project is also expressed in terms of Berry phase.
This part of my research program will be developed on simple models that are easy to manipulate and verify. Then if successful it will be extended to ab-initio codes.

[1] Notice that in case of static self-energies, namely without dephasing effects, the coupling between Berry phase and NEGF is possible, and this is the formulation I used in the non-linear optics

2. Theory of light controlled magnetism

Laser technology has grown explosively until reaching laser duration of few femto-seconds (fs), leading to a new era of non-equilibrium condensed-matter physics. One of the most intriguing applications is the use of ultra-short laser pulses to control the magnetization.[11, 12] How fast can the magnetization be changed? What are the fundamental and practical limits of magnetic writing and reading with light pulses? The need for fast non-volatile memories pushes the research of faster alternative approaches to write and read information. The answers to these questions will have great consequences on the future of data storage. This is why femto-magnetism promises the realization of a new generation of electronic devices and, at the same time, opens the opportunity to explore new emergent physics.

In magnetic memories, logical bits are stored by setting the magnetization vector of individual magnetic domains either up or down. However with the present technology, based on the application of external magnetic fields or spin-polarized currents, switching cannot be realized on time scales shorter than few picoseconds. A possible solution to overcome this limit and reach the THz regime is the use of ultra-short laser pulses, which last few femto-seconds. Indeed, after the first experimental demonstration that the magnetization can be controlled on the fs time-scale a very intense research activity has started.[11–13] This strong technological interest calls for advanced theoretical modeling. Nowadays semi-conductor companies invest a considerable amount of R&D money in modeling. Among the tools, ab-initio simulations are growing in importance, given their unique predictive power which can, in some cases, even replace the experimental investigation. Nowadays this is true for many aspects of modern electronic devices, but light-induced magnetization is still out of reach for ab-initio simulations. Besides the technological interest, a more fascinating aspect of femto-magnetism is that the main mechanisms behind the evolution of the magnetization are still not clear. In this project we will develop a theoretical framework able to clarify the physics behind femto-magnetism.

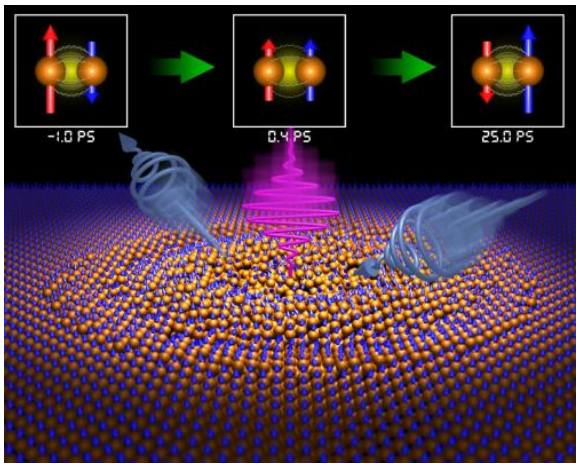

Figure 2.1: Schematic representation of light-induced magnetization.

2.1 State of the art and motivations

Different mechanisms have been proposed to explain why and how spin would evolve following a fs laser-pulse: the Inverse Faraday Effect, Electron-magnon scattering and Elliott-Yafet(EY) relaxation and so on.[14, 15] These mechanisms are often competing and rely on different physical phenomena. It is then important to have a precise estimate of their strength and time scale to understand the underlying physics of femto-second magnetization. In fact different experiments aimed at proving or disproving specific mechanisms have given opposite conclusions. As a consequence parameter-free modeling techniques have gained importance, receiving more and more attention in recent years. Ab-initio approaches used up to now are based on Time-Dependent Density Functional Theory (TD-DFT) and have focused only on specific physical processes, such as the role of the spin-orbit coupling (SOC) and the description of the Inverse Faraday Effects.[16] As an alternative, parameter free approaches which rely on phenomenological equations, have been used to study the impact of super-diffusive currents. Noticeably the ab-initio description of scattering processes in femto-magnetization, such as the EY mechanism, has been limited to the estimate of some key parameters.
The main difficulty of performing ab-initio simulations is their very large computational requirements. This is why TD-DFT, within adiabatic and local density approximations, was mainly used. However TD-DFT is not the correct theoretical framework, since it does not allow to include dissipative mechanisms, which are clearly needed. Coupling the electronic dynamics with the nuclei motion (Erenfest dynamics) is not a solution either since, on the fs time-scale, the atoms barely move. On the opposite side, an ab-initio Boltzmann equation could capture dissipative processes, but would not capture the coherent interaction with the laser pulse. The sole possible path to describe femto-magnetism is then to look for other approaches. With the advent of super-computers, there has been a substantial push to tackle the challenge of more demanding simulations. This makes possible to consider the ab-initio implementation of new and more sophisticated approaches. The description of femto-magnetization can be achieved within many-body perturbation theory, and in particular its non-equilibrium formulation based on Green functions. Non-equilibrium Green functions (NEGF) can describe dissipative mechanisms by means of the many body self-energy and capture the coherent interaction with the laser pulse on the same ground. The problem to extend this approach to the real-time magnetization lies in the fact that the magnetization is the sum two terms: the spin and the orbital magnetization. While the first can be easily calculated by studying the spin populations the second is difficult to estimate in periodic systems. From the experimental point of view there are indications that in these short time phenomena orbital magnetization can play a role comparable to the spin one. For this reason it is extremely import to have a method able to describe both components at the same level. Some authors have proposed a way to calculate orbital magnetization[17], but their approach is limited to equilibrium situations. In this project I aim to extend magnetization theory to non-equilibrium regime in such a way to capture the full magnetization dynamics.

2.2 Scientific goals

Theoretical, experimental and computational developments are three crucial instruments for the evolution of our understanding of physical phenomena. The key role of the latter, which emerged in recent years, is witnessed by the large number of publications and free available codes produced in recent years. It is then natural to aim at extending the ab-initio landscape to magnetization. In this project I would like to reach the two ambitious objectives:

- provide a description of the processes involved in light-induced magnetization and demagnetization and clarify their relative importance, by means of atomistic simulations.
- boost technological development of new magnetic devices, by suggesting specific design for new experiments

2.3 Organization and implementation of the project

The core of this project is an ab-initio description of light induced magnetization. We would like to capture all relevant physical ingredients needed to describe ultra-fast magnetization in pump and probe experiments. I will implement magnetization in periodic systems, and will include this implementation in the Lumen code. Lumen is an ab-initio code to study non-linear response of solids that includes corrections effects derived from many-body perturbation theory (MBPT). In the code correlation effects are limited to the ones described by static self-energies, but this approximation is enough to get accurate description of excitons in bulk materials. Orbital magnetization will be formulated in terms of wave-function phase, and this implementation will allow us to go beyond the linear regime. Notice that Lumen already includes the possibility to excite materials with polarized light and supports spin-orbit coupling.
Once we will have a description of magnetization in term of Berry phase we will be able to describe the interaction with the pump pulse and the subsequent coherent evolution of the magnetization. Dissipative process will be later included in by means of non-equilibrium Green functions. This part of the project will benefit from the merging of Green functions theory and Berry phase described in part I of the present research program,
The Spin-orbit coupling will be fully taken in to account in all steps by the underlying ab-initio atomistic simulation. The approach will be tested on specific materials. In particular I will consider two pilot computational-experiments on prototype materials which already have a well established experimental counterpart:

- the ultra-fast magnetization in Ni and $CoPt_3$ [18]
- light-induced demagnetization[19]

An approach able to describe these two cases will be likely general enough to provide an useful tool for the understanding of other ultra-fast magnetization and demagnetization processes. Moreover ab-initio simulations will be integrated with simple effective models (Hubbard and others) that offer an alternative way to study light-induced de/magnetization in presence of strong correlation. This part will be in collaboration with Andres Saul at CINaM, who has large expertise on lattice models for magnetic systems. These models will help to improve the ab-initio approaches based on MBPT.

2.4 Expected results

Hereafter the list of results expected from part II of my research program:

- Magnetization from first principles. Our developments will allow the description of light induced magnetization/demagnetization by means of numerical simulations including correlation effects within Many-body perturbation theory.
- Ab-initio packages freely distributed (GPL license) are nowadays a key tool for many research projects. The available software, especially for condensed matter physics, is mostly limited to equilibrium or steady state properties. The software developed in this project will be the first able to fully address this non-equilibrium physics. It will make feasible new computational experiments, which are presently not possible with any other software, neither open source nor commercially available.
- Understanding of opto-magnetic interaction from a parameter free Hamiltonian. Most of the understanding of femto-magnetism is based on intuitive physical concepts and model Hamiltonians. I would like to provide quantitative simulations based on a parameter free Hamiltonian that will allow us to clarify many of the mechanisms at the basis of light-induced magnetization. This approach will allow also to derive simplified Hamiltonian parametrized on Density Functional Theory.

- Design of opto-magnetic based devices made faster. Optimization of ultra-fast magnetic devices is only possible by theoretical understanding. However the understanding of light-induced magnetization is still strongly debated. The new insight provided by the successful realization of this project will help technological development.

2.5 Feasibility

The computational and theoretical developments will proceed in parallel. A first-principles implementation able to describe the time evolution of magnetic systems constitutes the core part of the project. This goal will be split in three work-packages: [A] theoretical development, [B] software development and [C] numerical simulations. The latter will be focused on the pilot experiments mentioned in the project that will serve to give feedback to both [A] and [B] during the developments. I underline that, despite being strongly interconnected, the activity of all three work-packages will start independently from the beginning of the project, thanks to the availability of the already working code, Lumen. Finally I will dedicate part of the activity to the exploration of simple models, parametrized from ab-initio simulations, and this part will in large part independent from the other tasks. Models will also help in analyzing the results of [C]. When we will have a working implementations of real-time magnetization we will investigate how correlation effects modify real-time dynamics and we will compare the results with simple solvable models. These two part will be done in collaboration with Andres Saul. Finally there will be an important work on code parallelization, and a part of our work will be devoted to the distribution of the calculations of correlations effects in real-time simulations. This part will be done in collaboration with Alexandre Zappelli a research engineer working at CINaM, expert in code optimization and parallelization.

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