Scientific report LabEx PALM 2015-2017

Laboratoire d'excellence



Physique : Atomes Lumière Matière

Table of contents

Introduction	5
 Focus Topic 1 "Quantum Matter: from the elementary to the strongly correlated"	9 12 13 14 15 16 16
 Focus Topic 2 "Complex systems: from out of equilibrium systems to biological matter"	21 of
 Focus Topic 3 "Ultrafast Dynamics: from radiation sources to multiscale responses"	29 31 33 35
 Topic 4 "Emergence, Evolution, Rapid Reaction" 1. New Materials 2. Biosciences 3. Plasma Chemistry 4. Free Radicals and Molecules Spectroscopy 5. Advanced Instrumentation 6. Exploratory Projects 	39 42 45 45 45
Topic 5 "Higher Education"1. PhD programs2. Master programs3. Popularization of physics	47 48
Topic 6 "Innovation and Technology Transfer"	52 52 53

Introduction

CONTEXT

This report covers the first 6 years of existence of LabEx PALM (2011 – 2019). The first scientific council was held two years ago, in February 2015, and acted as a highly valuable trigger for the preparation of the midterm evaluation by ANR. The outcome of the 2015 ANR evaluation was very positive and the LabEx has since then pursued its activities along the planned tracks at the end of the first 4 years. Initially scheduled for March 2018, the next evaluation from ANR will take place late spring 2018 after the evaluation of the IdEx Paris-Saclay. Our LabEx will apply for its continuation for 5 additional years after the end of 2019, a possibility which came out last February. Meanwhile the Paris-Saclay organization landscape has evolved and will keep evolving a lot until the 2020 horizon. Research departments have been launched with marginal funding mainly aimed at building up their structures and organizing their communities, and a COMUE (community of universities and engineering schools) has been created. A special attention was paid to complementarity to avoid duplicate actions with PhOM (Physics of Waves and Matter) research department or Paris-Saclay larger scale actions. The impact on the LabEx operating mode, organized around scientific projects, was minimal and will be pointed out where appropriate. In conclusion, in the new scheme of two poles Paris-Saclay and New UNI around Ecole Polytechnique, PALM appears as a well-suited tool to foster scientific interactions in our community.

ORGANIZATION OF THE REPORT

This report is divided into three sections: (i) an **activity report** focusing on scientific, innovation and higher education projects supported by our LabEx with subsection corresponding to the topics listed below. It has not the aim of encompassing all the science developed by the 750 researchers of the LabEx but to provide a good taste of the major funded scientific projects with key publications extracted from the list available at the end of each subsection and some highlights. Much more information can be found on the PALM website <u>https://www.LabEx-PALM.fr/</u> which has been enriched in the last 3 years with highlight contributions from the project PI's, still under completion; (ii) a shorter report about **future scientific prospects** for 2020-2025; (iii) an **appendix** with details about governance, participating labs and workforce, statistics, extended lists of funded projects, tables, graphs, budgets and a few statistics...

CONTOUR

The Scientific contour of PALM encompasses the physics of condensed matter, atomic and molecular physics, optics, lasers and "extreme" light, statistical physics and physical chemistry. The LabEx concentrates on three major topics whose focus have been revisited at the mid-term stage. Details about their evolution and their scientific extension are given in the introduction of the subsections corresponding to each topic:

- 1. Quantum Matter: from the elementary to the strongly correlated
- 2. Complex systems: from out equilibrium systems to biological matter
- 3. Ultrafast dynamics: from radiation sources to multi-scale responses

Aside from these three focus topics, a fourth topic named "Emergence, Evolution and Rapid Reaction" allows a rapid response to smaller scale projects and offers the possibility of a seed funding to emergent science or ideas in an open-minded spirit. Part of the physics at the interface with biology came to maturity and migrated to focus topic 2 which was redefined after the midterm review.

The two other topics link our LabEx with the surrounding world, academic through the higher education projects and socio economic through innovation and outreach actions. The topic innovation is run in common with LabEx NanoSaclay.

BUDGET

The budget figures by year are given below. PALM being originally designed not to be a long lasting project, it had been decided to allocate a higher share of the budget during the first phase (2012-2015) and a maximum at the beginning of each phase (i.e. in 2013 and 2016) to launch decisive actions while still keeping a sizeable part for the remaining time for fewer high - level projects.



OPERATION

The central operating tool of PALM is based on annual calls for proposals covering the six topics with a well-defined budget for each topic. This budget is approved upstream by the 11 institutions where the 35 partner labs come from. A collaborative character between two labs is required in the projects. This acts as a virtuous filter encouraging local collaborations, which has led in many cases to multi-partner in-PALM publications and results in structuring actions of the PALM community. The large-scale of the LabEx - ~750 researchers are affiliated to PALM- is for sure a key-point for enabling both this collaborating scheme and keeping a large diversity of scientific directions. This allows a real bottom-up spirit to define innovative projects.

Scientific excellence, novelty, structuring and collaboration are keywords in the evaluation of the projects. The topic boards have in charge the evaluation of the proposals, which are reviewed, depending on the amount requested, by the researchers of the board or by external referees (generally two). The topics boards and the steering committee which makes the final decision after the evaluation from the topic boards have been partially renewed after the mid-term evaluation. This procedure seems to satisfy the community, with a 4 months-delay between project submission and the funds availlability, and a \sim 30% success rate (ratio of the allocated budget to the resquested one).

The scientific actions of the annual call are listed and commented below, the project financial scale has extended for 2015-2017 from a few k€ to 160 k€:

- Equipment: multi-partner with leverage action for high budgets, exceptionally a single partner-project for a seed funding equipment on novel scientific directions.
- Senior chairs to attract prestigious researchers with conferences, courses, workshop organizations. They have been in
 operation until the start, in 2015, of a similar Paris-Saclay d'Alembert program, encompassing all the research spectrum
 of the IdEx.
- Junior chairs: aimed either at attracting high level young researchers in the idea of stabilizing them on the short term on a permanent position (external junior chair) or accelerating the initial carrier for recently hired young researchers and preparing them for obtaining further grants such as ANR-jeune or ERC (internal junior chair).
- Post doctoral grants: funding is granted only for a 2-years stay occurring in a 4 years time-slot after the PhD. Co-funding is possible.
- PhD fellowships: fully funded by the LabEx for a 3 years time: this program has been steadily increased since 2015, but the call was disrupted in 2017 because the financial rules of ANR prohibited to commit money beyond 2019. This restriction has now been lifted and coming calls now allow cofounding in view of the restricted budget planned for the next 2-years of the LabEx.
- Innnovation projects: they lie quite upstream, at the maturation level, in the chain leading from discoveries to patents. The focus is put on state-of-the-art instrumentation, innovative set-ups developed in the frame of the scientific activity, which are precious by-products which can fulfill academic or industrial requirements for other application fields. The PALM action, led together with the LabEx NanoSaclay, is complementary to those led by IDEX structures such as SATT dedicated to support technological innovation and the launching of start-ups. The innovation projets can fund personal costs, or small equipment.

It is important to note that these tools occur in a multiscale context of funding. First, PALM topics are also central in the research department PhOM (*Physics of Light and Matter*) of the University Paris-Saclay. Among the 7 different working groups structuring

the scientific perimeters of that research department, four of them, *Coherence and Quantum Correlations, Diluted or Ionised Matter: Plasma Atom and Molecule, Complex Systems and Matter* and the *Theory and Simulation* groups strongly resonate with our focus topics. This strong connection existing between PALM and PhOM has materialized in a series of successful actions that have been funded by the Idex: during the last couple of years several projects have been prepared to respond to large collaborative initiatives (IRS, *Initiatives de Recherche Stratégiques* de l'Idex) : our topics, in a joint action with the PhOM department have supported several of them. Among those projects, 3 were funded or at least labelled and partly funded, and they have seen a significant implication of PALM researchers:

- IQUPS about quantum technologies
- NAN'EAU a multimicroscopy appro; ach to study the dynamical behaviour of nano-objects in the liquid phase and in electrochemistry;
- PSI2, the Paris-Saclay International Programs for Physical Sciences and their Interfaces that is going to cover the gap of program for basic science before the proper functioning of the programs of the Institut Pascal, a program aimed at inviting scientists on given topics, similarly to Kavli institute. Several researchers of topic 1 are active members of the executive program committee of this initiative.

With a few hundreds k€ funding over 4 years for a broad community of researchers, these projects complement and extend the action of projects already funded by PALM with ambitions such as participating to European flagships (e.g. Quantum Technologies) or funding experimental platforms. At the regional level, heavy equipments are supported by Sesame grants now presented by the COMUE Paris-Saclay (encompassing all research domains). A regional network has been launched in 2017 on Quantum Technologies. Our ANR national funding agency supports collaborative projects at the national level. TheEquipEx Attolab, CilEx which are at the heart of PALM science have been early on funded by a special funding program (PIA, programme d'investissment d'avenir) and now come to operation, in close relation with our topic 3 for Attolab and CilEx (see report) etc..

Some other clear leverage effects can be identified among the 24 ERC grants obtained by our researchers (C. Pépin, topic 1; B. Gallet, topic 2; F. Quéré, topic 3) or CNRS awards such as silver medals (B. Dubrulle, topic 2) or Bonus Quality Research actions from Paris-Sud University (A. Santander, topic 1), to cite a few. Altogether with the chairs, PALM has therefore a clear role of **research accelerator**.

A total of 78 research projects has been funded in the period 2015-2017 (total of 145 since 2011) with an average of 56 k€ per research project over the entire life of the LabEx. This document is dedicated to exclusively report on **PALM-funded activity** and

the number of yearly publications –acknowledging PALM which is constantly repeated to project leaders but not completely integrated- seems to have come to a steady state, see graph.

Importantly, PALM strikes a balance between equipment and personnel expenditures at variance with most LabEx at the national level. In 2014, the national average was 55% for personal cost, whereas for PALM it is 35%.

Besides our pure research projects there are important channels which contribute to the **international visibility** at all levels **from confirmed researchers to undergraduate students**:



- an on-demand visitors program for stays up to 3 months (18 visitors in 2015-2017 from 11 countries);
- the funding of workshops and schools organized by members of PALM, 42 on the 2015-2017 period with a financial support up to 5 k€);
- the full funding of one "PALM" school per focus topic, which will be held in 2018-2019;
- the diffusion of PALM -based science with outreach and financial support for the creation of innovative practical labs at the Master level and financial support for Master internships;
- a web-site in French and English not only for intra-LabEx use but now rather aimed at publicizing our action both at the researchers and general public level. The website has been considerably enriched by highlights on a large number of projects since 2015.
- last but not least a **popularization** section on our web-site which has been one of the priorities of the LabEx in the last 2 years.

Some of these actions linking research and education have also been the seed for a thinktank leading to a much broader graduate school proposal PHYSICS@UPSaclay in the context of the call launched in 2017.

Finally **intra PALM community actions** have been entailed to animate our community with a few Newsletters, PhOM research seminars whose organizers are commonly PALM members and some were co-funded. There were topical days on quantum Information, Complex Systems, UltraFast technical developments, Theory.

GENDER ISSUE

26% of PALM researchers, 24% of leaders of funded project are women, with a 35% participation ratio in our scientific and higher education committees. 38% of post-doctorant fellowships and 30% of PhD fellowships have been allocated to women. Beyond these statistics regarding gender issue, the promotion of women in PALM has been encouraged by supporting them around their maternity leave, e.g. by supporting grants for non-permanent reasercheers to avoid any disruption in their high level research. This is explicitely written in our annual call for research projects.

As for the current PALM day which will be part of the evalution by the committee, 3 out of 8 speakers are women presenting some of our emblematic project for years 2015-2017

About references cited in the report

All references of published papers **acknowledging** funding from **PALM** are listed at the end of the scientific report and sorted by name of the first author. They are cited as [NAME of first author YEAR] in the text of the report.

References to other papers either from PALM members or not appear as footnotes.

Focus Topic 1 "Quantum Matter: from the elementary to the strongly correlated"

A significant recent trend in physics has been the recognition of the common ground between the atomic molecular and optical (AMO) physics community on the one hand, and the condensed matter physics community on the other. Quantum information processing, pioneered using quantum optics techniques (trapped atoms and ions, single photons) have found analogs in condensed matter systems such as superconducting circuits, and quantum dots. These systems constitute "artificial atoms" which promise to improve the scalability of quantum information processing setups. On the other hand, questions concerning strong correlations, entanglement and coherence, which have long been of importance to the condensed matter community, are now also being addressed using ultracold atoms manipulated by laser beams. This development holds the hope of better understanding systems such as high temperature superconductors. The field of quantum transport, in which matter waves (either electrons or atoms) propagate under the influence of various forces, either in a solid or an optical potential, also constitutes a new area of common ground between the two communities.

The Quantum Matter focus topic (1) encourages researchers in the LabEx PALM to explore these subjects and we try especially hard to encourage collaborations and synergies between condensed matter and AMO physics. One of the most striking examples is the use of optical lattices to simulate spin Hamiltonians such as the Ising model [LABUHN 2016]. In PALM, at the Laboratoire Charles Fabry, there has been a successful effort to use programmable arrays of microscopic single atom traps to realize such simulations. Loading these traps with Rydberg atoms provides a system with strong, long range and anisotropic interactions. This specific project was not funded by PALM, but we make a brief mention of it here because, this achievement opens the way to simulations of exotic matter. We expect much more progress on this topic in the near future.

The LabEx PALM did fund several other projects addressing condensed matter issues using cold atoms. Another area of overlap was the use of circuit QED physics to realize artificial atoms and networks. Many other topics in correlated systems were also pursued: topics in high temperature superconductors, two-dimensional electron gases, frustrated magnetism and ferroelectrics. The sections below contain an overview of some of the larger projects which were funded by the LabEx.

Another role of the LabEx is to help in the establishment of versatile technical platforms which can benefit many different scientific projects. A good example is the NanoFIBHel project. This project was initiated by three groups at the C2N, the CEA-SPEC and the LPS laboratories, and permitted the purchase of a focused helium ion beam for state of the art ion beam lithography. It represents an outlay of more than 1 MEuros, which were provided by several different organizations. The initial investment was made by the PALM LabEx after which several others (LabEx NanoSaclay, INP (CNRS), CEA, Lidex Nanodesign, and DIM NanoK) followed suit. Another technological platform created at the SPEC laboratory to assemble multilevel heterostructures (the ZerHall project).

See Highlight 1 (project NanoFIBHEL by Richard Deblock) and Highlight 2 (project ZerHall by François Parmentier)

1. Correlated systems of cold atoms

With the demonstration of cooling methods to achieve quantum degeneracy atom-atom correlations have been present in most experiments. Solving problems in systems with a large number of interacting quantum particles - manybody problems - is notoriously challenging. Sometimes both numerical and analytical theoretical methods fail to provide a satisfying answer. An alternative approach consists in finding an experimental situation which can simulate a many-body problem and use its results to gain insights applicable to other systems.

The past decade has seen great progress in the control of individual quantum objects like trapped ions, superconducting qubits and neutral or Rydberg atoms. All these platforms are capable of measuring spatial distributions and correlations between individual particles. But some paradigmatic manifestations of many-body effects are difficult to see in spatial correlations, and multi-particle correlations between other degrees of freedom play a fundamental role. In the CORUM project (collaboration between the LCF and the LPTMS), a new approach is being developed to investigate other degrees of freedom by measuring the momentum distribution of individual particles in guantum gases. The work exploits the properties of Helium-4 atoms brought to quantum degeneracy in a metastable state. In particular, the large internal energy of metastable Helium-4 yields the unique possibility to detect individual atoms in three-dimensions after a long time of free-fall with Micro-Channel Plates (MCP, see figure (a)). To benchmark the capabilities of this technique, a Bose-Einstein condensate is loaded into a three-dimensional crystal of light of relatively large amplitude and measurements are made of the momentum distribution (see figure (b)). In this situation, the celebrated Bose-Hubbard Hamiltonian can be numerically solved with a Quantum Monte-Carlo approach (QMC). The measured distributions perfectly match the QMC calculations. This work opens a new route to investigate interacting lattice systems through momentum correlations.

Highlight 1. Nanofabrication with a Focused Helium Ion Beam R. Deblock, A. Kasumov, R. Weil, (LPS), P.F Orfila, D. Estève (SPEC), D. Mailly (C2N).

The investigation of quantum matter at nanometer length scales has made tremendous progress during these last decades. In this context, three teams Phynano (C2N), Quantronics (SPEC) and Meso (LPS), have combined their efforts and expertise in nanofabrication, in order to create a new platform entirely devoted to nanofabrication beyond the ten nanometer scale. This platform, unique in France, open to the UPSaclay community, has been recently installed in the C2N laboratory. This platform is based on a recently acquired Orion Nanofab focused ion beam microscope manufactured by Zeiss.



The Orion Nanofab microscope is based on a helium and a neon field ion gun. The ultimate probe size in a scanning electron microscope (SEM) is limited by diffraction and chromatic aberration. Due to the very high source brightness and the shorter wavelength of the helium ions, it is possible to focus the ion beam to a smaller probe size relative to the SEM. The helium ion beam is capable of sub-10 nm nanofabrication as well as high resolution imaging capability in the same instrument. On the other hand the neon ion beam offers precise machining and nanofabrication capabilities due to higher sputter yields in ion beam milling and faster resists exposure in ion beam lithography. The ORION NanoFab multiple beam solution permits direct write ion machining from the meso-scale provided by neon to the nanometer scale made possible by helium.

Gas injection systems (GIS) are commonly employed on SEM and FIB tools. The use of the beam to induce local chemical reactions on a substrate allows direct writing of nanostructures (either metallic or insulating) without the added pattern transfer steps that lithography requires. The ORION NanoFab helium ion microscope (HIM) is equipped with such a system. The structures can be observed and measured using the same beam which does not create any lateral ion damage in contrast with the Ga ion source.

The instrument has been in service since 2016. The researchers have been working on the optimization of the machine in terms of resolution and reproducibility of the etching conditions on various materials. Some experiments have been performed: realization of nano-slits and holes in suspended graphene, nanogaps in metallic electrodes and deposition of metallic nanoelectrodes using the injection system. The nanowires exhibit superconductivity below 6K and can be used as superconducting contacts on mesoscopic samples.

Results achieved in the framework of the project NanoFIBHEL funded by topic 1 and carried out by Richard Deblock (LPS) and Daniel Estève (SPEC).



CORUM project. a. diagram of apparatus, b. 3D plot of experimental data showing diffraction peaks and the wings of their distributions

Key publication

H. Cayla, C. Carcy, Q. Bouton, R. Chang, G. Carleo, M. Mancini, D. Clément, "Single-atom-resolved probing of lattice gases in momentum space, arXiv 1710.08392

The presence of atom-atom correlations has required revisiting some traditional AMO techniques as simple as the observation of light scattering from an atomic cloud. When the cloud is very dense and the frequency of the light is tuned near an atomic resonance, light induced electric dipoles are large and interact via the dipole-dipole interactions, which in 10

turn modify the scattering. This phenomenon may ultimately affect the accuracy of atom-based sensors such as atomic clocks. Many experiments have been devoted to the study of scattering by dilute cold laser-cooled atomic samples, but the case of high density has received much less attention. In an initial experiment in this regime, performed in 2014 at the Laboratoire Charles Fabry using a dense, microscopic cloud of cold rubidium atoms, a state-of-the art theoretical model failed to reproduce the data, most likely due to the complicated internal structure of the atoms.

The ECONOMIQUE project (collaboration between the LCF and the LAC) financed the PhD thesis of Stephan Jennewein. The project consisted again in measuring the optical response of a cloud (see figure a), with the innovation of applying a strong magnetic field (300 G) to isolate a two-level structure in rubidium (figure b). Measurements were made of the transmission of a probe propagating through the cloud for different atom numbers (from 5 to 100, figure c). The team developed a new model based on the generalization of the Bloch-Maxwell formalism, and found a much better agreement with the data than any previous studies. For the first time a theory is able to fit experimental results in this high-density regime. This opens new perspectives to realize recent proposals, such as a mirror consisting of a plane of atoms structured in an array, or non-

linearities induced by the interactions that could provide new ways to generate non-classical states of light.

Key publication

S. Jennewein, M. Besbes, N.J. Schilder, S.D. Jenkins, C. Sauvan, J. Ruostekoski, J.J. Greffet, Y.R.P. Sortais, A. Browaeys, *Coherent Scattering of Near-Resonant Light by a Dense Microscopic Cold Atomic Cloud*, Physical Review Letters, 116 (2016)



ECONOMIQUE project. a. Diagram of experimental apparatus, b. Atomic level scheme, c. Transmission vs laser detuning for different atom numbers

Highlight 2. Layer assembly technique for the realization of ultra-clean graphene van der Waals heterostructures

P. Brasseur, M. Jo, M. Seo, P. Roulleau, F. D. Parmentier (SPEC), T. Taniguchi, K. Watanabe (NIMS, Japan).

In order to access the very peculiar fundamental electronic properties of graphene, such as the *quantum Hall ferromagnetism* arising at large magnetic field due to the presence of both *spin* and *valley* symmetries [1], it is crucial to realize graphene samples of the highest quality. A major impedement comes from the contamination of the graphene surface during sample processing, *e.g.* by polymers used for electron beam lithography. To prevent this, and obtain the highest possible sample qualities, we have implemented in SPEC a *van der Waals* heterostructures assembly technique first developed in 2013 [2], where the graphene crystal is encapsulated between two atomically flat hexagonal boron nitride (h-BN) crystals. This encapsulation allows processing the samples (*e.g.* depositing metallic contacts connecting the edges of the graphene crystal [2]) without polluting the graphene surface, yielding ultra-high quality devices with record mobilities.



The assembly platform implemented in SPEC allows constructing clean, multilayer heterostructures, with micrometer alignement precision. In particular, we are able to realize state-of-the-art samples where few-nanometers-thick graphite flakes are incorporated in the heterostructure to be used as electrostatic gates, so as to tune the carrier density in graphene. Panel (a) of the above figure shows the low temperature transport characteristics of such a bilayer graphene device, where the derivative of the sample resistance with respect to back gate voltage V_{gate} is plotted versus V_{gate} and applied magnetic field. The yellow lines emphasize the quantum Hall effect states at filling factors 3, 2, 1 (dot-dashed line), and the antiferrmoagnetic state at filling factor 0 (dotted line).

Those four last states demonstrate the very high quality of the device. Using this platform, we are able to fabricate complex multiterminal devices (panel (b)), with various types of metallic electrodes, including superconductors. Panel (c) shows several multiterminal graphene devices (made from the same graphene flake) connected to Ta/Nb superconducting leads realizing flux loops (yellow dashed lines).

Using this state of the art fabrication technique, we will probe unexplored aspect o quantum transport in graphene, such as electron quantum optics, heat transport In the quantum Hall regime, and low temperature THz plasmonics.

M. O. Goerbig, *Electronic properties of graphene in a strong magnetic field*. Rev. Mod. Phys. 83, 1193 (2011) L. Wang, et al. *One-Dimensional Electrical Contact to a Two-Dimensional Material*. Science 342, 614-617 (2013)

Results achieved in the framework of the project ZerHall funded by topic 1 and carried out by François Parmentier (SPEC).

2. Circuit quantum electrodynamics

Among the most basic questions in guantum electronic transport is what happens when a guantum conductor is driven by an ac voltage. The QECmicroscOP project (collaboration between Lab. de Physique des Solides and SPEC) recently addressed theoretically what happens when this ac voltage is itself truly quantum mechanical in nature (being described by for example a squeezed state or a Fock state). Existing systems might realize such a situation: by coupling a tunnel junction to a superconducting microwave cavity. It was shown that the current of the conductor has signatures that directly reflect the non-classical nature of the driving microwaves. In that sense, a measure of an electric current can be used as a detector of the quantum nature of the microwave electric field. Such a feature is illustrated in the figure showing a Fock state with two microwave photons in the cavity which entails a strongly non-monotonic dependence of the differential conductance of the tunnel junction with the bias voltage. Moreover, the transport is naturally associated with quasi-probabilities that can become negative. The figure below shows the differential conductance dl/dV versus dc bias voltage V for a tunnel junction coupled to a microwave cavity initially prepared in the Fock state n=2. The striking signature of a non-classical state here is the strongly non-monotonic dependence of the first few conductance plateaus on voltage; in particular, the height of the second plateau (h2) is smaller than the first (h1). This is in one to one correspondence in the Fock state with exactly two photons in the cavity.



Key publication

J.-R. Souquet, M.J. Woolley, J. Gabelli, P. Simon and A.A. Clerk, *Photon assisted tunnelling with non classical light*, Nature Communications, 5, 5562 (2014)

The quantum electrodynamics of circuits also was also an important feature of the QuantumClimat project (CPhT IPhT, LPS and LPTMS). Here the properties of topological Bloch bands were investigated. One of the main manifestations of such properties is the presence of protected edge states. For non-interacting fermions this leads to the celebrated integer quantum Hall effect and to novel conducting surface states of topological insulators and semi-metals. These properties can also be accessed with bosonic systems such as cold atoms, photons and polaritons.

The Su-Schrieffer-Heeger (SSH) model defined on the dimerized one-dimensional lattice with two sites per unit cell is one of the simplest models demonstrating topological properties; The edge states are protected by the bipartite nature of the system (particles can hop only between the two sublattices).

The researchers studied topology in the strong coupling regime of quantum circuits in which the rotating wave approximation (RWA) is not applicable, leading to the appearance of counter rotating (pairing) terms. Such strong coupling limit also leads to the evolution of the Jaynes-Cummings model towards the Rabi model when describing the qubit-cavity interaction, and to the super radiant phase in Dicke model with a macroscopic number of photons in the ground state but has not been studied in he framework of topological systems.

The winding of the topological Zak phase across the Brillouin zone can be measured by a reflection measurement of (microwave) light. Our method probes bulk quantities and can be implemented even in small systems (~10 unit cells). Figures (c-d) show the imaginary part of the reflection coefficient as a function of the light frequency ω for the trivial phase (ω <v, figure c) and the topological phase (ω >u, figure d). In the two phases the spectrum revealed by the resonances is the same. However the envelope function of the resonances amplitudes show the winding of the Zak phase. In the trivial phase, the envelope has the same value at the bottom and top of the bands. In the topological phase the envelope of the bands is a monotonically increasing function.



Emergence of topological Bogoliubov bosonic excitations in the relatively strong coupling limit of an LC (inductance-capacitance) onedimensional quantum circuit

T. Goren, K. Plekhanov, F. Appas, K. Le Hur, *Topological Zak Phase in Strongly-Coupled LC Circuits*, Submitted (2017)

Quantum optics experiments were also supported by PALM. A Schrödinger cat state is a superposition of two coherent states with opposite phases, and is an example with which one can implement a quantum gate. The main challenge is to be able to generate them efficiently. The HAQI project (LCF and LAC) has demonstrated that such a cat state can be produced by the "fusion" of two photons: two singlephoton states are mixed on a symmetric beamsplitter, leading to the photon coalescence effect, and a homodyne measurement projects the state onto a cat state. This operation can be iterated, increasing the complexity of the generated state, and this could be performed efficiently with a reliable quantum memory.

Key publication

J. Etesse, M. Bouillard, B. Kanseri, and R. Tualle-Brouri, *Experimental Generation of Squeezed Cat States with an Operation Allowing Iterative Growth*, Phys. Rev. Lett. 114, 193602 (2017)

3. High temperature superconductors

The concept of symmetries governing the behaviour of physical states is maybe the most robust in theoretical physics. From the formation of nuclei to the Higgs-Boson the symmetries are fundamental in the determination of every emerging state in high energy physics. It would be quite remarkable if a phenomenon as complex as high temperature superconductivity would be governed as well by an overall emergent symmetry. Suggestions about the existence of a pseudo-spin symmetry in the background of the physics of superconducting cuprates have been introduced since the early days of these compounds and has been revived over the years in different contexts. In all cases, the main idea is that one can rotate the d-wave superconducting state towards another state of matter degenerate in energy. The physics is then controlled solely by the powerful constraint of the emergent symmetry. A vast region of the phase diagram is dominated by fluctuations between those states. Here, an emerging SU(2) symmetry connecting the d-wave superconducting state with a d-wave charge order is the main ingredient of the physics of superconducting cuprates.

This emergent SU(2) symmetry allows one to understand the origin of the mysterious pseudo-gap phase out of which superconductivity emerges. In the EXCELCIUS project (LLB and IPhT) researchers have given a new theoretical account of the physics of the pseudo-gap phase in terms of the emergence of local patches of particle-hole pairs generated by SU(2) symmetry fluctuations. The proliferation of these local patches accounts naturally for the robustness of the pseudo-gap phase to disturbances like disorder or magnetic field and is shown to gap out part of the Fermi surface, leading to the formation of the Fermi arcs.

A recent idea was put forward suggesting that nano-size electrical loop currents, travelling around tiny circuits of only three atoms each, are formed in the pseudo-gap phase. These loop currents can be viewed as the hallmark of a ferro-toroidal order, breaking both time reversal and parity symmetries, while preserving the lattice translation invariance. In the EXCELSCIUS project the experiments attempted to track down features predicted by this intriguing model, such as the magnetic field that the loop currents must generate. One expects in each crystalline unit cell two loop currents flowing in opposite directions, producing two antiparallel moments. The difficulty here is that the fields, and the size of the loops that generate them, are tiny. Measuring such fields is, however, exactly what neutrons are extremely good at. Like tiny compass needles, they explore the magnetic field on length scales going down to atomic dimensions. Magnetic signals more than 3 orders of magnitude weaker than the dominant nuclear part could be measured on the instruments 4F1 (LLB) and D7 (ILL) using method of polarized neutron diffraction. The the measurements allowed not only to determine with precision the magnitude of the tiny moments but equally their direction and the extent to which they are correlated in space, and all of this could be followed as a function of temperature. These results form an eagerly awaited stringent set of conditions on any model that tries to describe the physics of the pseudo-gap phase. They provide us with a way better idea of the form that the postulated current loops may take. In particular, as a function of temperature, loop currents could evolve from a classical to a quantum regime, with a quantum superposition a several loop current patterns.

Key publications

X. Montiel, T. Kloss, C. Pépin, *Local particle-hole pair excitations by SU(2) symmetry fluctuations*, Scientific Reports 7, 3477 (2017)

L. Mangin-Thro, Y. Sidis, A. Wildes & P. Bourges, Intra-unit-cell magnetic correlations near optimal doping in YBa2Cu3O6.85, Nature Communications 6, 7705 (2015)

Bismuth cuprate superconductors were also studied in another project, FemtoARC (collaboration between LSI, LPS and Soleil). In this project, angle resolved photoemission (ARPES) techniques were used to study non-thermal distributions of cooper pairs. This research is discussed further in the next highlight.

See Highlight 3 (project FemtoARC by Luca Perfetti)

Highlight 3. Filling of a supercondonducting gap by photoinduced fluctuations C. Piovera, K. Van der Beek, L. Perfetti (LSI), E. Papalazarou, M. Marsi (LPS), Z. Zhang, M. d'Astuto (IMPMC), A. Taleb-Ibrahimi (Synchrotron SOLEIL).

Unlike conventional superconductors, the cuprates display a layered structure and a pairing interaction extending over a few lattice sites. As a consequence the d-wave gap does not follow a mean-field behaviour. As shown by the figure, the thermal excitation (right panel) or photoexcitation (left panel) gradually fill the gapped spectral region. By employing time and angle resolved photoelectron spectroscopy we observed that thermal fluctuations are effective only for temperatures near the critical value whereas photoinduced fluctuations scale linearly at low pumping fluence. The peculiar behavior of the photoexcited state indicates that Cooper pairs scatter with a nonthermal distribution of low-energy excitations. We show that an intermediate coupling model accounting for the finite pair breaking explains the gap filling both in the near-nodal as well as in the off-nodal direction.



Z. Zhang, C. Piovera, E. Papalazarou, M. Marsi, M. d'Astuto, C. J. van der Beek, A. Taleb-Ibrahimi and L. Perfetti, *Photoinduced filling of near-nodal gap in Bi*₂Sr₂CaCu₂O₈₊₅, Physical Review B 96, 064510 (2017)





EXCELSCIUS project A. Temperature dependence of the polarized neutron magnetic scattering intensity. B. staggered orbital moments induced by loop currents. C. Schematic representation of the magnetic moment deduced from the polarized neutron experiment. At high temperature (starting from room temperature), the moment points perpendicular to the expected loop current whereas a tilt occurs at low temperature (down to 100 K) when the moment strengthens. All measurements are done above the superconducting temperature (Tc=89 K)

4. Two dimensional electron gases in correlated oxides

Two-dimensional electron gases (2DEGs) in transition metal oxides (TMOs) can show metal-to-insulator transitions, superconductivity, magnetism, or spin-polarized states, and are thus an active field of research promising for applications semiconductor technology. beyond However, their fabrication and use are hampered by the need to grow complex oxide layers using complicated and expensive techniques. In the framework of the ELECTROX project (CSNSM, LPS, Soleil), researchers developed a new, simple, and essentially universal method to fabricate 2DEGs in functional oxides. It consists in the deposition of a thin layer (a few Å) of an elementary reducing agent, such as pure AI, over the surface of an oxide in vacuum. By a redox reaction, the reducing agent pumps oxygen from the substrate, oxidizes into an insulating passivation layer, and simultaneously dopes the first atomic planes of the underlying oxide, thus forming a pristine 2DEG. The quantum-well states of such a 2DEG, for instance in SrTiO3 -a non-magnetic bulk-insulating TMO that is considered to be a prime candidate platform for oxide electronics, are then directly measured using angle-resolved photoemission spectroscopy (ARPES).

Using this idea in a follow up project, 2DEG2USE, the team successfully created a 2DEG at the interface between EuO -a ferromagnetic insulator- and SrTiO3. This functional interface was created through the thermal evopartion, simply using a Knudsen cell, of pure metallic Eu on SrTiO3. The redox reaction reduces the SrTiO3 surface, thereby creating a metallic 2DEG and a EuO overlayer. ARPES was used to probe the electronic structure of the interface, and magnetization measurements to confirm the ferromagnetic character of the EuO overlayer. Depending on its thickness, the overlayer can be either paramagnetic (1ML) or ferromagnetic (2ML) as shown by the in-plane magnetization curves in figure below (a).



2DEG2USE project a. magnetization curves of a 2D electron gas. b, c electronic structure of 2D electron gases for 1 or 2 monolayers

Key publications

T. C. Rödel, F. Fortuna, S. Sengupta, E. Frantzeskakis, P. Le Fèvre, F. Bertran, B. Mercey, S. Matzen, G. Agnus, T. Maroutian, P. Lecoeur, A. F. Santander-Syro, *Universal Fabrication of 2D Electron Systems in Functional Oxides*, Advanced Materials 28, 1976 (2016)

P. Lömker, T. C. Rödel, T. Gerber, F. Fortuna, E. Frantzeskakis, P. Le Fèvre, F. Bertran, M. Müller, A. F. Santander-Syro, *Two-dimensional electron system at the magnetically tunable EuO/SrTiO3 interface*, Physical Review Materials, 1(6) 062001 (2017)

The electronic structure of the corresponding 2DEG is shown in figures (b) and (c) when the overlayer thickness is, respectively, 1ML and 2ML. The parabolic electronic states correspond to surface-confined electrons that induce the metallic character of the interface.

The results open new perspectives for investigating the interaction of the well-known 2DEG on SrTiO3 with magnetism, and prove that the new device-friendly growth technique can be used to enhance the functionalities of future all-oxide devices.

5. Electronic properties and topology in 3D quantum materials

Transport, whether of current, heat, spin or some other physical quantity is an essential occupation of condensed matter physics. Several Quantum Matter projects were devoted to this extremely broad topic.

A collaboration between theorists at the LPS and the LPTMS in the MAGNSOCS project has investigated quantum oscillations in small-gap insulators. Quantum oscillations are a well-known technique in the study of electron systems in the presence of a magnetic field. They provide valuable information about the band structure of metallic systems by mapping out their Fermi surface. According to the generally accepted picture, the energy bands get partially quantized by the magnetic field into Landau levels, and oscillations occur every time the Fermi energy crosses a Landau level. It comes therefore as a huge surprise that small-gap insulators can, under certain circumstances, also display quantum oscillations in the absence of a Fermi surface. In order to understand this unusual situation one needs to distinguish two classes of guantum oscillations. While those due to the pure Fermi surface, such as the Shubnikov-de Haas oscillations in the magneto-resistance, are absent in an insulator that has no quantum states available at the Fermi level, a second class of quantum oscillations requires taking into account the full Fermi sea, i.e. all states up to the Fermi level. Such a situation is for example encountered in de Haas-van Alphen oscillations in the orbital magnetization of an electron system. In this case, also an insulator can display quantum oscillations if the gap is locally inverted, e.g. when the conduction and valence bands overlap and are hybridized such that the valence band upper edge has the shape of a circle and plays the role of an effective Fermi surface.

Key publication

H. K. Pal, F. Piéchon, J.-N. Fuchs, M. O. Goerbig, G. Montambaux, *Chemical potential asymmetry and quantum oscillations in insulators*, Physics Review B 94, 125140 (2016)

The Joseph Bismuth project (LPS and ISMO) was devoted to using the current phase relationship in a superconducting junction to probe conduction in topological insulators. The current through such materials is predicted to run only through a few perfectly ballistic channels, called topological edge states. The topological insulator candidate is a monocrystalline bismuth nanowire whose crystalline orientation is chosen such that it contains two topological surfaces, each with one-dimensional edge states. The researchers found that the supercurrent through a 1.4 micrometer-long monocrystalline bismuth nanowire has a sawtooth-shaped dependence on the phase difference between the superconductors at its ends. This indicates ballistic rather than diffusive transport. The fact that transport is ballistic over such a long distance hints to a possible topological protection against scattering in those wires [Murani 2017].

In the PIRITIS project (LPS and LSI) researchers introduced a novel experiment based on inducing controlled disorder in prototype materials like Bi₂Se₃, Bi₂Te₃, and Bi₂Te₂Se by charged particle irradiation. Stable charged point defects in the bulk of layered topological insulators has proven useful for achieving topological compounds that are really insulating in their bulk. In order to study the energymomentum dispersion of Dirac electrons in a doped topological insulator, researchers from LPS made use of an experimental approach based on angle-resolved photoemission spectroscopy. They demonstrated that irradiation with electron beams up to 2.5 MeV compensates intrinsic charged defects, thus bringing the Fermi level back into the bulk gap and reaching the charge neutrality point. Controlling the beam fluence, they could tune bulk conductivity from p- (hole-like) to n-type (electron-like), crossing the Dirac point and back, while preserving the Dirac energy dispersion [Caputo 2016].

Spin orbit coupling and disorder was the topic the SOC project (LPS and Soleil). The compound Sr₂IrO₄ is considered as the archetype of a novel type of Mott insulator, the "Spin Orbit Mott Insulator. While it should be metallic, since Ir has 5 electrons in its 5d shell, it is an antiferromagnetic insulator. This cannot be understood in the standard Mott scenario because the electronic correlations, responsible for Mott insulating behavior, are relatively small for 5d transition metals. However, it was shown that the Spin-Orbit Coupling, which is very strong for heavy atoms like Ir, induces a reconstruction of the electronic structure with a narrow, half-filled Jeff=1/2 band, where even small Coulomb repulsion may open a gap. This situation bears similarities to cuprates. The researchers synthesized Sr₂(Ir_{1-x}Rh_x)O₄, Rh being a 4d element isovalent to Ir. Using ARPES, they observed a metallic state which does not present quasiparticle peaks, even at the optimal doping, the bands are not renormalized compared to the band calculations, and a "`pseudogap" of about 30meV is present on the entire Fermi surface. This is attributed to an incoherent behavior resulting from the interplay between correlation and disorder introduced by Rh (publication in preparation). The effect of disorder in a strongly correlated compound is a rich theoretical and experimental subject. This study of the Insulator-metal transition in an exotic Mott insulator brings a novel point of view in this large field of research.

The topic of disorder has also been of increasing interest in cold atom research. Although no projects on disorder in cold atoms were funded by PALM between 2013 and 2017, a group at the LCF has been using optical speckle to produce a disordered potential in which atoms with coherence lengths of several microns can travel (or be localized.) Evidence of Anderson localization in a three dimensional disordered potential was observed at the LCF in 2012¹. In more recent work a demonstration of weak localization, in the form of a coherent backscattering peak. was performed in 2015². Future work will be directed towards observing and locating the mobility edge in a three dimensional disorder and in observing the phenomenon of coherent Forward scattering, another signature of Anderson localization.

6. Frustrated Rare Earth Pyrochores

In the context of classical spins, 3 dimensional Pyrochlores ($R_2B_2O_7$, R = rare-earth; B = non-magnetic transition element) with their highly frustrated corner-sharing lattice are a gold mine for stabilizing many unconventional magnetic states, such as spin-ices, spin liquids or exotic ordered phases. Because of the rare earth atoms (R), the stabilization of one given phase comes from the compromise between single ion anisotropy, dipolar interactions and magnetic exchange. While titanates (B=Ti) have been largely investigated in the last 20 years, stannates (B=Sn) open new possibilities. When dipolar interactions dominate, a specific "Palmer-Chalker" state with a first order transition

¹ F. Jendrzejewski, A. Bernard, K. Muller, P. Cheinet, V. Josse, M. Piraud, L. Pezzé, L. Sanchez-Palencia, A. Aspect, and P. Bouyer, Nat. Phys. **8**, 398 (2012).

was predicted long ago: the exploration of Er₂Sn₂O₇ (project 1DMag, LLB-SPEC) using sub-K neutron scattering and magnetization experiments clearly established the first evidence for such a transition. A strange persistence of dynamics, a landmark observation of frustration in many highly frustrated antiferromagnets has yet to be interpreted [Petit 2017].

Just surprising as for classical spins, quantum spin liquids can also be stabilized in some rare earth pyrochlores. Here the low energy physics of Kramers-protected doublets maps onto effective spin ½ systems. The recently funded QUANTUMPYROMAN project (LPS, LLB and ICMMO) has proposed two challenging avenues in that vein (i) an in depth investigation of the fluctuating state of the Yb₂Ti₂O₇ compound, a QSL candidate, and (ii) exploration of the physics of the new pyrochlore magnet Sm₂Zr₂O₇. Indeed, the zirconate (B=Zr) family has been largely overlooked up to now, in part because of difficulties related to their synthesis and might be the seed for novel exotic phases.



Neutron evidence for a magnetic transition at 108 mK in Er₂Sn₂O₇

7. Ferroelectrics

In the ECO project (LPS and LLB), researchers made permittivity, and electric polarization dielectric measurements along with an accurate description of the structural and microscopic magnetic properties obtained on NdMn₂O₅ from high resolution x-ray and neutron diffraction studies performed on large facilities and at the LPS. NdMn₂O₅ is shown to be ferroelectric, although the magnitude of the polarization is much weaker than that of the other multiferroic members. The direction of the polarization is along the crystallographic b axis and its magnitude can be tuned with the application of a magnetic field. However, unlike the other multiferroic members of this series, the ferroelectricity in NdMn2O5 emerges in an incommensurate magnetic state. The study also provides evidence in support of the influence of the rare-earth size on the magnetoelectric phase diagram.

² K. Muller, J. Richard, V. V. Volchkov, V. Denechaud, P. Bouyer, A. Aspect, and V. Josse, Phys. Rev. Lett., **114**, 205301 (2015).

Key publications

S. Chattopadhyay, V. Balédent, F. Damay, A. Gukasov, E. Moshopoulou, P. Auban-Senzier, C. Pasquier, G. André, F. Porcher, E. Elkaim, C. Doubrovsky, M. Greenblatt, and P. Foury-Leylekian, *Evidence of incommensurate multiferroicity in NdMn*₂O₅, Physical Review B 93, 104406 (2016)

Exotic magnetic phenomena were also investigated in the CHIRAL project (collaboration between the LLB and Soleil).

Using x-ray studies the group was able to demonstrate the existence of a chiral magnet characterized by an invariant lattice parameter. The results are discussed further in the highlight below.

See Highlight 4 (project CHIRAL by Isabelle Mirebeau)



ECO project Left: Powder neutron diffraction pattern and magnetic structure of NdMn2O5. Right: Electric polarization measured by depolarisation current

Highlight 4. 'Invar' behavior in a chiral helimagnet

N. Martin, I. Mirebeau (LLB), M. Deutsch (Université de Lorraine, CRM2), J.-P. Itié, J.-P. Rueff (Synchrotron SOLEIL), U.K. Rössler, K. Koepernik (TU Dresden, Germany), L.N. Fomicheva, A.V. Tsvyashchenko (Institute for High Pressure Physics, Moscow).

The history of 'invar' behavior goes back to the end of XIXth century, when C.E. Guillaume synthetized and studied Fe-Ni ferromagnetic alloys, characterized by an invariant lattice parameter around room temperature (hence the term 'invar'). Since its discovery, several models have been developped to understand this peculiar phenomenon, and magneto-elastic coupling is unanimously acknowledged as an essential ingredient, explaining why different properties of the materials, such as the atomic volume V, the local magnetic moment m or the bulk modulus B can be affected. One of the most popular theories of the 'invar' effect is due to P. Weiss who introduced the so-called 2 γ -model, where a high-spin (HS) state with large atomic volume and magnetic moment competes with a low-spin (LS) metastable state with reduced volume and moment. Here, a thermal activation of the LS state counteracts lattice thermal expansion (LTE). However, as pointed out by D.I. Khomskii and F.V. Kusmartsev, this simple mechanism cannot explain the cancelation of LTE over large temperature intervals as experimentally observed. It is therefore necessary to feed additional ingredients into the model such as intersite elastic coupling between minority spin states, and verify their validity on real systems. In this context, chiral magnet MnGe appears as a good playground since neutron powder diffraction (NPD) has revealed a HS-LS transition at low temperature, within its helimagnetic state.



Here, we have made use of X-ray powder diffraction (XPD) and emission spectroscopy (XES) to study the pressure-dependence of the unit cell volume V (Fig. a) and Mn local moment m (Fig. b) of MnGe. Our initial aim was to ascertain the existence of an intermediate LS state at T = 300 K, i.e. in the paramagnetic regime where long-range magnetic order is absent. As seen in Fig. a, V describes a remarkable hysteresis loop upon pressure cycling. This loop opens upon decompression at pC1 \approx 7 GPa, which corresponds fairly well to the HS-LS critical pressure of \approx 6 GPa determined by NPD. Interestingly, the existence of such elastic irreversibilities perfectly supports the idea of Khomski and Kumartsev that the nucleation of low volume LS regions induces local strains which couple over finite distances through the sample. In other words, they build up a macroscopic energy barrier that has to be overcome to transform the system back to its initial HS state. We have developped a thermodynamic model of the spin-lattice transition which accounts for the observed V = f(p) curve. In parallel, a challenging XES experiment has allowed us to follow the pressure-dependence of the local Mn moment up to \approx 40 GPa. As shown in Fig. b, m collapses at pC2 \approx 23 GPa and leads to a non-magnetic state. This is another way of verifying the existence of an intermediate LS state and assessing its metastable nature.

In summary, our study establishes MnGe as the first known example of 'invar' chiral magnet. Moreover, our findings directly verify the theoretical prediction of KK that intersite coupling must play an important role in the appearance of 'invar' anomalies, thus updating common viewpoints on the physics at play in these systems.

N. Martin, M. Deutsch, J.-P. Itié, J.-P. Rueff, U.K. Rössler, K. Koepernik, L.N. Fomicheva, A.V. Tsvyashchenko and I. Mirebeau, *Magnetovolume effect, macroscopic hysteresis, and moment collapse in the paramagnetic state of cubic MnGe under pressure*, Physical Review B 93, 214404 (2016)

Results achieved in the framework of the project CHIRAL funded by topic 1 and carried out by Isabelle Mirebeau (LLB).

Focus Topic 2 "Complex systems: from out of equilibrium systems to biological matter"

The current focus topic 2 of PALM " Complex systems: from out of equilibrium systems to biological matter" gathers experimentalists, theoreticians and numerical modellers working on various objects (glasses, granular media, turbulent fluids, soft matter, fracture problems, algorithms,...). Statistical physics of systems at equilibrium provides today a well-established framework to classical thermodynamics. However, most of the systems encountered in condensed matter physics and beyond, e.g. in biological, industrial or natural systems, are out of equilibrium, either because of the presence of an external forcing or because they cannot relax back to equilibrium. The objects of interest come from multiple domains: spin or structural glasses, granular and soft matter, simple or complex fluids, biological or active matter, lasers. They raise questions dealing with fracture, interfaces, suspensions, instabilities, chaos, turbulence, mixing, networks, and information. However, the approaches used to address these problems are common and pertain to statistical physics. Understanding the emergent dynamics in out-of-equilibrium systems represents one of the major challenges for the next decade because they are ubiquitous and cannot be understood simply through small modifications of equilibrium physics. Moreover, they are directly connected to important societal needs such as energy, climate, environment or health. The Paris-Saclay campus gathers an important number of international level teams working in distinct subfields where these issues play a major role. One of the main challenges we want to face is making these various communities, working on different objects, talk to each other, and establish a common conceptual framework. This transdisciplinary project, with theoretical, numerical and experimental aspects, will structure this community around the following most significant domains, in closed relationship with education and some important industrial problems and societal questions, with an important potential of investment return.

Most of the systems we meet in Nature display some sort of structure or organisation. It is then interesting to understand how those structures arise, and how they evolve with time. Though it is now fairly well understood why a structure arises in systems at equilibrium, self-organization emerging from the cooperative interactions among the constituents of a system maintained out-of-equilibrium is less easily understood, but very often this system displays properties that are different from those of the constituents. This question is ubiquitous in essence. Out-of-equilibrium self-organized systems appear in a variety of contexts: almost everybody has some experience with binary solutions with a positive enthalpy of mixing, liquid crystals, non-equilibrium patterns in laser-induced transformations, turbulence in chaotic systems, ...Many biological systems fall into this far-from-equilibrium category. Also, self-assembly or self-organisation of nanostructures is seen as a basis for many developments toward high-performance materials, components, and devices in nanotechnology and information technology.

At the mesoscale, experimental and computational models are developed to study self-organized systems in considerable detail: the evolution of these structures is the system response to an initial instability, and it may be described using free energy functions, generalized to account for medium heterogeneities or the existence of interfaces. Also, fluctuations are important and influence the structure and the properties of the system. When free energy functions do not exist, the description of the dynamics of those far-from-equilibrium systems must rely on different assumptions: for instance, in biochemical systems the starting point could be a reaction-diffusion equation, while in fluid dynamics the Navier-Stokes equation is often a reasonable starting point.

In an attempt to track the physical phenomena and study the emerging properties in complex systems, the researchers of focus topic 2 of PALM often use a mix of elements specific to the physics of equilibrium and far-from-equilibrium systems. These approaches are the core/common language of this community. In contrast with the fundamental differences and origins of these approaches, there are often similarities in their forms, and there are common basic elements that are needed to foster our understanding of the formation end evolution of these systems. Among these common elements, the need to define field variables (order parameters) for describing the spatial structure, the presence of interfaces that separate spatial domains, and the existence of defects.

See Highlight 5 (project HYPERPOL by Alberto Rosso)

While a truly unified picture of the problems of this branch of physics is still out-of-reach, the activities of focus topic 2 of PALM foster the development of the tools needed to analyse and understand the origins of the organisation of many diverse systems (Disordered and glassy systems, soft matter, active matter and biological systems, fluids and lasers instabilities, turbulence and mixing, ...). These tools are developed to discover the similarities observed in non-equilibrium and driven systems, and to establish bridges between the methods used to this purpose. This should help to reinforce a common conceptual framework and to create a dynamic between them around their respective challenges, relevant for their societal impact and in close relationship with training and education.

In the study of matter, both the living and the inanimate one, breakthrough discoveries can spring from the study of emergent behaviours, phenomena that root their existence in the interactions between many sub-units, but whose presence cannot be deeded by the detailed knowledge of the sub-units alone. Assembly of molecules and geometrically frustrated matter illustrate this notion, but also cognition is an emergent phenomenon and fluid instabilities or protein aggregation as well.

Highlight 5. Spin temperature and hyperpolarization in disordered magnets A. De Luca, I. Rodiguez-Arias, Alberto Rosso (LPTMS), M. Bauer (IphT)

Dynamic Nuclear Polarization (DNP) is to date the most effective technique to increase the nuclear polarization opening disruptive perspectives for medical applications. However, our understanding of the physical mechanisms that trigger hyperpolarization is still poor. In DNP, the nuclear spins are driven to a hyperpolarized state by microwave saturation of a collection of electron spins interacting with the nuclear spin system. A striking experimental evidence is the thermal mixing of the ensemble of different nuclear spins (13C, 15N, 89Y...): their enhanced polarizations are well described by a single spin temperature possibly one thousand time smaller than the temperature of the bath. But how can a quantum system appear thermal and colder when irradiated by microwaves? In which way the spin temperature can be controlled acting on the experimental parameters?



In our work we show that the spin temperature concept is directly connected to quantum ergodicity and that the resulting stationary state strongly depends on the ergodicity properties of the spin many-body eigenstates. In particular

dipolar interactions compete with disorder induced by local magnetic fields resulting in two distinct dynamical phases: while for weak interaction, only a small enhancement of polarization is observed, for strong interactions the spins collectively equilibrate to an extremely low effective temperature that boosts DNP efficiency. Our central finding is that the nuclear hyperpolarization increases steadily upon reducing the Dipolar strength. Interestingly, the highest polarization is reached at a point where the establishment of a spin temperature is just about to break down due to the incipient many-body localization transition in the electron spin system. A very recent experiment seems to confirm our main prediction (Bodenhausen group).

A. De Luca, I Rodriguez-Arias, M. Mueller, A. Rosso, *Thermalization and many-body localization in systems under dynamic nuclear polarization*, Physical Review B 94, 014203 (2016)

Results achieved within the project HYPERPOL funded by topic 2 and carried out by Alberto Rosso (LPTMS) and Michel Bauer (IPhT).

In this respect, the discussion of the developments of this axis is organised around the general topic of emergent phenomena and it is illustrated by specific classes of materials and system-related problems. Below, we provide some examples detailing how PALM has brought new talents, expertise, and resources to the initiatives of topic 2, as a result of funded collaborative work. These examples illustrate how the impact of the LabEx is well beyond the simple scientific production over the period covered by this report, and it has created real lasting synergies between our laboratories

1. Synthetic review of the projects

The ordering of the following description of the funded projects the topics follows an arbitrary loose progression of length scales, from the atomic scale to large scale aggregates of biomolecules and fluids. In all cases, the focus of the discussion is on the character of emergence for the particular system, the role of research approaches for these problems, and the effort to identify the organising principles ruling these phenomena.

The project DYNINTDISQUANT started in 2015 by Marco Schirò (IPhT & E. Bogomolny of LPTMS) is a junior chair (2015-2018) that is still in progress. M. Schirò has been recently hired at CNRS in IPhT and the focus of this research project is on the study of interacting and disordered quantum systems that can display rich emerging dynamical phenomena far from thermal eauilibrium. Their understanding represents a challenging frontier of research at the interface between statistical physics and condensed matter [Lupo 2016]. Schirò's project aims at exploring this novel research direction building upon the expertise in the field of out equilibrium quantum dynamics of IPhT and LPTMS. The budget of this project (140k€) has covered the costs for a two-year postdoctoral position.

Key publication

S. J. Thomson & M. Schiro, *Time Evolution of Many-Body Localized Systems with the Flow Equation Approach* (2017)

The project SWEET, SWift Engineered Equilibration or Thermalization, started in 2016, is carried out by E. Trizac (LPTMS, Alexei Chepelianskii of LPS & O. Dulieu, LAC) and involves the funding of a post-doctorant fellow. The project aims at studying the relaxation time, an intrinsic property of any system, be it natural or artificial, which indicates how long the equilibrium recovery takes after a sudden change of a control parameter. Physical laws in principle allow accelerating dramatically such a thermalization process, taking advantage of an appropriate driving of the system. This is the rationale behind the recently introduced shortcut to adiabaticity protocols (STA) that are the focus of this project. STA ideas have hitherto been worked out and demonstrated for isolated quantum and classical systems only. The extension to open systems in contact with a thermostat is a stumbling block for a wealth of applications. The project tries enlarging the scope of this idea, and address a typical soft matter system, colloids in an optical trap, creating a confining potential, the properties of which can be controlled in time. Beyond its fundamental interest, the SWEET method paves the way for applications in micro and nano devices, but also to monitor mesoscopic chemical or biological processes, where the role of thermal fluctuations cannot be neglected.

Key publication

A. Le Cunuder, I.A. Martinez, A. Petrosyan, D. Guery-Odelin, E. Trizac, S. Ciliberto, *Fast equilibrium switch of a micro mechanical oscillator*, Applied Physics Letters, 109, 5 (2016)

Two grants (post-doc grants) were awarded to L. Zdeborovà in 2013 [TASC] and 2016 [SAMURAI] for a total amount of 180k€. The main focus of the Tasc project deals with the theory and applications of a concept known as "spatial coupling". This concept has emerged to be a crucial ingredient of future signal processing units, error correcting technologies and other inference-like problems of large practical interest. It is known that inference problems are closely related to some statistical physics models. Also, in many such problems the onset of algorithmic hardness can be understood via the appearance of a metastable state that blocks the dynamics of the algorithm. The main idea behind spatial coupling is to design the problem in such a way this metastability is avoided via nucleation of the equilibrium state that corresponds to the signal to be inferred. The perspectives of spatial coupling are far-reaching, its theoretical understanding is still poor. The TASC project fills this gap by analysing the nucleation-like dynamics of spatially coupled inference problems [Caltagirone 2014]. The other project, Samurai, deals with the unsupervised learning of representations of data, stands behind the recent progress in machine learning, and paves the way towards artificial intelligence [Lesieur 2017]. Representation learning can be modelled as a particular kind of a mean-field spin glass. Methods stemming from physics of disordered systems turn out as very instrumental in solving and understanding this model. The simplest (replica symmetric) version of these methods applied to this model and it has already led to very interesting results. The SAMURAI project tries to analyse an extension called replica-symmetry breaking (RSB), as done recently by S. Franz for a related problem of perceptron. The main focus of the project that involves a two-year post-doc is on two interesting cases of representation learning - clustering of high-dimensional data and dictionary learning. L. Zdeborovà has also obtained an ERC starting grant in 2016, on the statistical mechanics of learning.

The project MUSA (2017) by G. Foffi (LPS & S. Franz of LPTMS) has funded a post-doctorant position. The project aims at using the theoretical and computational tools of statistical mechanics and soft matter to rationalize the study of self-assembly, that is the guiding principle according to which synthetic building blocks spontaneously assemble to form structures with a higher level of complexity. Mastering self-assembly is of great fundamental and applied importance, therefore it occupies a solid position in research worldwide. In soft matter, the possibility to engineer particles with ever increasing complexity as building blocks for selfassembly has rejuvenated the field, but the complexity of the syntheses, their characterization and throughput can be a limiting factor. To overcome this shortcoming, G. Foffi proposes to take a fresh look to the problem of Multiselective Self-Assembly (MuSA). Here, complexity naturally emerges from the combination of several very simple objects and not by tinkering them individually. The key advantage is that simple building blocks mean a more robust, flexible and reproducible route to self-assembly. The key points studied in this project concern the study of the self-assembly of selectively interactive particles, the stability theory for

multicomponent mixtures with selective interactions, and the glass transition in systems with selective interactions.

The project Emersion 2015 by G. Baldinozzi (SPMS & V. Pontikis of LSI) principally covers the fabrication costs of layered strcutures that are necessary to obtain an experimental benchmark for theory and numerical modelling [Pontikis 2017]. The object of this project is the numerical modelling and the direct observation of the effects related to interactions between defects at ultra-thin coherent or semicoherent out-of-equilibrium interfaces between metals with a positive enthalpy of mixing. The understanding the physical effects coming from the competition of short and medium-range interactions on the atomic mixing at the nanoscale is aimed at a better comprehension of this phenomenon and, in the end, at exploiting those metastable architectured interfaces with the exciting long-term perspective of obtaining better properties for technological systems like coating and thermal barriers, especially when these systems are maintained out-of-equilibrium by the effects of temperature and/or irradiation.

The project POPS (2016) by C. Poulard (LPS & P. Cortet at FAST) is a collaboration between the two partners that involves the funding of a two-year post-doctorant. POPS put focus on the modelling of the adhesion mechanisms of soft materials on a substrate. This is a longstanding problem which remain largely open nowadays. It is motivated by the industrial importance of potential progresses in the control of pressure sensitive adhesives, but also by the fact it constitutes a very rich fundamental problem at the frontier between rheology, fracture mechanics and polymer physics. The project POPS aims at exploring the physics of adhesion using patterned substrate design and, in particular, at using these substrates as a tool to tune the balance between the different mechanisms allowing for adhesion such as viscous dissipation and elastic hysteresis.

The project DRYORDER (2015) by C. Rountree (SPEC & G. Gauthier of FAST) is about how the drying mechanisms of a thin layer of colloidal suspension yields a solid film with varying porosity [Rountree 2017], optical transparency, and resistance to fracture. These macroscopic properties are the signature of nanoscopic organization and nature of the chemical bonds within the porous media. The objective within the DRYORDER project is to study the influence of the drying rate, film thickness, and bead dimension and polydispersity on the solid obtained from the drying of model colloidal suspensions. The two partners have defined an innovative experimental protocol combining experimental mechanics (macroscale) measurements and atomic force microscopy imaging (nanoscale) and they have revealed a selection the film non-trivial of structure (crystalline/amorphous) by the drying rate. DRYORDER funds a PhD candidate to address the long-standing problem of understanding how macroscopic properties emerge from the nanoscopic structure.

The project BINGFLOW (2017) by L. Talon (FAST & A. Rosso of LPTMS) funds a post-doctorant. The object of this project is the investigation of the flows of yield stress

materials, fluids that require a minimal amount of stress for flowing in porous media. If yield stress fluid are encountered in many applications, it remains that the characterization of the flow in disordered systems is still a challenging and controversial task. The main difficulty arises from the strong interplay between the non-linear rheology and the disorder of the system. In particular, close to the flowing critical pressure, the flow correlation exhibits a fat tail distribution reminiscent to critical systems in disorder media. The main focus is the study of the viscoplastic flow in highly heterogeneous porous media. In particular, the generalization of Darcy's law for more heterogeneous media and the invasion front of a Newtonian fluid in porous media. Thanks to the different expertises of this collaboration, these problems are investigated by the means of different approaches: numerical simulations, statistical physics and asymptotic expansions.

The project FLRAM (2017) by C. Nardini (SPEC & K Mallick IphT) is a junior chair project aiming at funding a postdoctorant fellow to help developing the computational aspects. The last few years have seen an upsurge of studies at the boundary between biology and physics. A clear example is 'active matter', defined as those media where non-thermal energy is locally transformed in the presence of dissipation into an effective motion. It comprises very diverse systems from bacteria, catalytic colloidal particles and vibrated rods to animal flocks. Understanding their collective behavior is a fundamental question with the exciting long-term perspective of long-term technological applications such as the creation of self-assembly materials. The aim of this project is to unveil the role of fluctuations and long-range interactions on the collective behavior of active matter, combining analytical and computational tools [Stenhammar 2017]. While both of them have been mainly overlooked in the theoretical literature, they are expected to play a fundamental role. Indeed, most experiments comprise only a small number of active particles and long-range interactions due to hydrodynamic or phoretic effects are often present.

Key publication

J. Stenhammar, C. Nardini, R.W. Nash, D. Marenduzzo, A. Morozov, *Role of Correlations in the Collective Behavior of Microswimmer Suspensions*, Physical Review Letters, 119 (2017)

The project PERCEFOULE (2017) by C. Appert-Rolland (LPT & A. Nicolas of LPTMS) is a small project about the study of the movement of rather dense crowds starting from experimental measurements using cameras adapted for the study of dense populations of individuals. The project involves the actual collection of data and its modelling to establish an analysis of the collective dynamics under controlled situations.

Also related to the junior chair program, several other projects funded since 2016 illustrate the successful opening toward the interface between physics and biological systems. The funded junior chair MIPOBIO [Bittner 2016] and project PHYSBIOPS-a reflect this inflection and confirm the increased involvement of topic 2 in this thematic area. Focus

topic 2 of PALM was instrumental in supporting the Physics/Biology interface through the "Physique des Systèmes Biologiques Paris- Saclay" (PhysBioPS) project.

See Highlight 6 (project PhysBioPS by Martin Lenz)

Coordinated by four researchers from four different laboratories, this initiative federates the Physics/Biology interface within PALM and more broadly the Paris-Saclay Campus. It organizes a seminar series entirely funded by PALM, workshops typically co-funded by other institutions, as well as a biannual international conference. With its 14 participating laboratories, PhysBioPS has been the main unifying effort of the Physics/Biology interface within Paris-Saclay since its inception in late 2012. Since 2015, this effort at the junction of physics and biology has now gained enough momentum to be included and funded within the boundaries of our topic. Also, contacts are developed with the different colleagues involved in this research line. The corresponding biophysics activity is often multi scale (from molecules such as nucleic acids or proteins, to network and then cells, tissues or populations). It is furthermore intrinsically out of equilibrium, with a dynamical response emerging in a broad range of time scales. Progress in this field relies both on original theoretical ideas, and on the development of new experimental techniques, with some important overlap with the communities of soft matter physics, already well represented within focus topic 2. It therefore appeared both timely and scientifically natural to foster the presence of topic 2 in this activity, favouring the structuration of several small and scattered activities within a large number of laboratories of the Paris-Saclay Campus.

Highlight 6. Unifying the physics-biology interface in Paris-Saclay

G. Foffi (LPS), A. Gautreau (Dpt. de Biologie, Ecole polytechnique), M. Lenz (LPTMS), C. Marlière (ISMO), N. Westbrook (LCF).

While widely represented in the Paris-Saclay area, the physics-biology interface there is very scattered geographically. To help structure it internally as well as bolster its national and international visibility, we organize three types of operations: a biennial international conference, regular topical workshops and a regular seminar series (see list of events below). As of late 2016 these events had brought together as event organizers researchers from 18 different institutes ("laboratoires") in the Paris-Saclay area, and participants from many more.



International Conference

3rd International Conference on Physics and Biological Systems (Oct. 2016) 2nd International Conference on Physics and Biological Systems (Jun. 2014)

Workshops

Physical chemistry of the cell 2017. (Nov. 2017) Modeling of 3D genome organization. (Oct. 2017) In honor of Françoise Livolant: Geometry & interactions in self-assembled biological systems. (Mar. 2017) Electroactivity of biological systems (Nov. 2015) Physical chemistry of the cell 2015 (Nov. 2015) Virophysics 2015, at the frontier of physics and virology (Jun. 2015) From Soft Matter to Cell Biology workshop (Jun. 2015) 2015 Paris-Saclay biophysics day (Jun. 2015) Biomechanics across scales workshop (Feb. 2015) Ultrafast spectroscopy for biologically relevant systems (Jan. 2015) Microfluidics for biology (PALM session, part of the microfluidics GDR) (Jul. 2014) New optical microscopies for biomedical applications (Sep. 2013) Physical approches to bacterial communities (Mar. 2013)

Seminar series

Physics-Biology Interface seminar series (every second week since 2012)

Results achieved within the project PhysBioPS funded by topic 2 and carried out by Martin Lenz (LPTMS), Giuseppe Foffi (LPS), Christian Marlière (ISMO), and Nathalie Westbrook (LCF).

Highlight 7. Energy transfers in rotating turbulence P.-P. Cortet, N. Machicoane, A. Campagne, F. Moisy (FAST), B. Gallet (SPEC).

Rotation of planets and stars is a key ingredient of the turbulent dynamics in geophysical and astrophysical flows. Yet a comprehensive understanding of the influence of a global rotation on turbulence has still not been achieved. This is particularly true in the regime of strong rotation where turbulence is dominated by the dynamics of inertial waves, the internal waves specific to rotating fluids, resulting from the restoring action of the Coriolis force.

In this project, we were interested in the fundamental question of the nature of the energy transfers in rotating turbulence, and particularly in the relevance of the weak turbulence theoretical framework to describe this state. We have developed a new rotating turbulence experiment in which the injected power could be directly measured. It consists in an impeller set in rotation in a water tank, the ensemble being itself driven under rotation by a precision rotating platform. We have demonstrated a strong rotation-induced "drag reduction", i.e. a reduction in the global dissipated power when rotation strengthens, which turns out to agree with the scaling law predicted by the weak turbulence formalism. However, measurements of the turbulent velocity fluctuations by particle image velocimetry suggested another interpretation to this drag reduction: it is actually the result of an inhibition of the turbulence related to the bidimensionnalization of the large cale vortices of the flow. This result suggests that, to approach experimentally the weak turbulence regime, it is crucial to inject energy preferentially in wavy modes rather than in vortical modes.

For even stronger background rotation, a new regime is observed: the quasi-geostrophic (vertically invariant) mean flow driven by the impeller rotation is subjected to "barotropic" azimuthal modulation. This modulation drives a relative velocity between the impeller blades and the mean flow, which leads to the excitation of a wake of inertial waves behind each blade, similar to the wake of surface waves behind a ship. We speculate that the nonlinear interaction of these wakes could form, at larger Reynolds number, a particular state of inertial wave turbulence.



(a) An impeller is set under rotation at a rate ω in a water-filled tank. The ensemble is driven under rotation by the "Gyroflow" rotating platform at a rate Ω . The torque Γ developed by the motor of the impeller is measured as well as the velocity field in the vertical green area. (b) Mean dissipated power $P=(\Gamma\omega)$ in the flow (in grey) normalized by its value P^{∞} without rotation as a function of the control Rossby number $Ro=\omega/\Omega$ (the upper branch corresponds to the anti-cyclonic case $\omega Q < 0$ and the lower branch to the cyclonic case $\omega Q > 0$). Colored markers correspond to the global dissipated power estimated from the measurements of the velocity fluctuations. The good agreement between the two data series shows that the transfers of energy still follow the scaling law for isotropic turbulence without rotation

N. Machicoane, F. Moisy, P.-P. Cortet, *Two-dimensionalization of the flow driven by a slowly rotating impeller in a rapidly rotating fluid*, Physical Review Fluids 1, 073701 (2016) A. Campagne, N. Machicoane, B. Gallet, P.-P. Cortet, F. Moisy, *Turbulent drag in a rotating frame*, Journal of Fluid Mechanics Rapids 794, R5

A. Campagne, N. Machicoane, B. Gallet, P.-P. Cortet, F. Moisy, Turbulent drag in a rotating frame, Journal of Fluid Mechanics Rapids 794, R5 (2016)

Results achieved within the project GEOTURB funded by topic 2 and carried out by Pierre-Philippe Cortet (FAST) and Paul Billant (LadHyX)

The project Interdist, funding the acquisition of 2 highresolution cameras, has started in 2016 and it is carried on by Francois Daviaud of SPEC in collaboration with FAST. Its main focus is on the dissipative scales in an experimental turbulent swirling flow by using Tomographic Particle Imagery Velocity (PIV) measurements and a new criterion for detection of dissipative structures based on the work of Duchon and Robert. Funding the acquisition of 2 new highresolution ScMos cameras completed the existing PIV system (used for instance in [Humbert 2017]). The 33 k \in of PALM were complemented by a CNRS grant concurring to the total cost of the project (75 k \in). The two high-resolution cameras being portable, they can be easily shared within the laboratories of PALM having similar interests for visualising turbulent flows (FAST and LADHYX in particular).

The project GEOTURB (2015) by P. Cortet (Fast & P. Billant Ladhyx) involves a partial funding of two post-doctorant fellows (see highlight #7). Rotating and stratified flows can support eddy and wave turbulence due to the existence of

anisotropic dispersive waves, named inertial and internal waves respectively [Campagne 2015, Campagne 2016, Machicoane 2016]. The project GEOTURB aims at addressing two key issues of geophysical turbulence: the direction and mechanisms of the energy cascade, and the relevance of the weak wave turbulence theory. These issues are addressed by means of numerical simulations of rotating/stratified turbulence, at laboratory LadHyX, and experiments of purely rotating turbulence, at laboratory FAST. The project capitalizes the experience of N. Machicoane post-doctorant at FAST that has started to setup the forcing device for the new rotating turbulence experiment.

See Highlight 7 (project GEOTURB by Pierre-Philippe Cortet)

2. Brief review of the main results obtained since 2015 of projects started during the first phase of PALM

SOFT2HARD: MECHANICAL PROPERTIES OF JAMMING SYSTEMS

Many soft matter systems undergo a transition from a liquidlike to a solid-like state upon the concentration of nano- or microscopic objects dispersed in a carrier fluid. The scientific object of this project is how exactly the systems "jam" towards the solid-like state, and how the organisation of the objects leads to emerging mechanical properties of the ensemble [Gaillard 2015, Ferreira 2017]. Four research teams at the LPS, FAST, and LPMC have joined together to investigate a range of systems which include drying or sedimenting nano- or microparticle dispersions, creaming emulsions and film-forming bacteria dispersions. Common to all systems is the fact that the concentration of the objects occurs at a "soft wall" imposed by the presence of a gas/liquid interface. In some cases, the presence of two gas/liquid interfaces leads to a "soft confinement". Understanding how soft systems become hard in the presence of such soft walls is the new research direction that can help understanding jamming systems. The spatial and temporal evolution of the dispersed systems in 3D and at high resolution is followed by this confocal microscope, installed at LPS but with access to other PALM members. The funds given PALM (180k€) has allowed leveraging similar amount within a Sesame Region Ile de France project. The microscope was inaugurated at the end of 2013 and several projects are now running in parallel (biofilm formation, structure and dye distribution of cholesteric droplets used as 3D microlasers, cellular dynamics, creaming emulsions...). Also, W. Drenckhan PI of this project has obtained an ERC grant.

Key publication

J. Ferreira, Interplay between bulk self-assembly, interfacial and foaming properties in a catanionic surfactant mixture of varying composition, Soft Matter 13, 7197-7206 (2017)

TURB&CRACKS: SCALE COUPLING & DISSIPATION IN HETEROGENEOUS FRACTURE & TURBULENCE

In both heterogeneous fracture and turbulence problems, the complexity stems from the separation between the (macroscopic) scale over which forcing is applied, and the (microscopic) scale where the dissipation occurs. As a result, the behaviour observed at the continuum-scale level is fully dominated by rare and extreme microscale events, the dynamics of which is coupled over a variety of time and length scales. The core of project Turb&Cracks lead by D. Bonamy was to provide time and space-resolved imaging of these complex couplings in highly controlled/highly modulated experiments. The acquisition of an ultra-fast camera has been integrated within the setups developed at SPCSI and SPEC to study fracture in disordered solids and turbulent flows, respectively [Piroird 2016, Barés accepted]. The funds given by PALM in 2013 (120k€) have also allowed significant leveraging: a CFR PhD student funded by CEA was obtained (2015-2018) and the results obtained within this project have set the basis for a consortium for responding to an ANR call.

Key publication

J. Barés, A. Dubois, M.L. Hattali, D. Dalmas, D. Bonamy, Aftershock sequences and seismic-like organization of acoustic events produced by a single propagating crack, Nature communication (accepted)

FLEXPAINTINGS: CRACK PATTERNS IN DRYING LAYERS

This project funded in 2013 (45k€) concerns the mechanical behaviour of complex thin-film systems undergoing external mechanical stresses. It analyses the effect of a deformable substrate on a layer in consolidation (colloidal and / or polymer dispersions) and the application to art paintings by linking the morphologies of fractures observed on a pictorial layer to the mechanical properties of the canvas and under layers of paint [Giorgiutti 2016]. A traction machine installed at FAST laboratory, allows controlling and measuring the deformation applied to the substrate (membranes) and linking the measurements to the morphologies of fractures that appear in the upper layer in consolidation. This small project has set the basis for a larger project named DEEPPAINT on the follow up of this subject funding a PhD in 2014-2017 involving LLB and FAST.



Crack patterns in some parts of Mona Lisa. A similar array of parallel cracks in a brittle colloidal layer (20 μ m thick) on a stretched sublayer (middle sketch) was investigated using the biaxial testing apparatus

See Highlight 8 (project DEEPPAINT by Frédérique Giorgiutti-Dauphiné)

CHAIRS

The senior chairs correspond to a consolidator grant. The junior chairs (internal or external) funded by topic 2 of PALM correspond to starting grant for young researchers who either got recently a permanent position in one of the PALM

laboratories or a grant for a group that is willing to hire a young scientist.

This mechanism has proven to be extremely successful as most of the external junior chairs funded between 2014 and 2017 have become internal at the moment of the actual start of the grant, because the selected candidate had obtained a permanent job in a laboratory of the LabEx. 4 chairs were funded between 2014 and 2017, and some of them are still in progress. Also, a couple of postdoctoral grants were funded along the same lines. In the following, a brief review of the results obtained since 2015 within the chair projects is presented.

The TURBONDES senior chair grant (142k€) shared among SPEC, FAST, and LadhyX has allowed inviting during the year 2014 Prof. S. Nazarenko of Warwick University (GB), a world recognized expert in the field of wave turbulence. It has enabled and structured many interactions among experimentalist and theoreticians of the Paris Saclay Campus. Among the related scientific production, it is worth noticing the results obtained by B. Gallet and B. Dubrulle (PI of this project) in the field of particle propagation. Also, following his stay, S. Nazarenko has applied in 2016 for a CNRS DR1 staff position in France and has strengthened his collaborations with the laboratories of PALM. We have to stress that since 2016 the senior chair program has been phased out as this kind of consolidator stays of senior scientists has been replaced by the d'Alembert program of Idex and the scientists invited within this new program do not appear in this report.

Key publication

E-W. Saw, D. Kuzzay, D. Faranda, A. Guittonneau, F. Daviaud, C. Wiertel-Gasquet, V. Padilla, B. Dubrulle, *Experimental characterization of extreme events of inertial dissipation in a turbulent swirling flow*, Nature Communication 7, 12466 (2016)

The chair BIOFIB (2013-2015), concerns M. Lenz (theoretician, hired in 2012 at LPTMS) and it deals with the characterization of robust and generic organizational principles for biofilaments on multiple scales (LPTMS-LPS). The object of the project is the study of cell mechanics [Lenz 2014, Chiaruttini 2015, Ronceray 2015, Grindy 2016, Ronceray 2016, Ruckert 2017] that relies on the geometrical organization of protein filaments, and its failure is related to diseases ranging from sickle cell anemia to Alzheimer's to cancer. Surprisingly, even in healthy cells the detailed morphology of these vital structures is very variable: the project aims at understanding this paradoxical combination of reliability and randomness in the cell's architecture, by theoretically characterize robust, generic organizational

Highlight 8. Crack opening : from colloidal systems to paintings M. Léang, F. Giorgiutti-Dauphiné, L. Pauchard (FAST)

Shrinkage cracks are observed in many materials, such as polymer coatings, colloidal systems and in paintings where great interest lies in deducing quantitative information on the material with the aim of proposing authentication methods. We have conducted experimental measurements on the crack opening induced by the drying of colloidal layers and compare these results to the case of a pictorial layer. We propose a simple model to predict the crack width as a function of the thickness of the drying layer, based on the balance between the drying stress buildup and the shear frictional stress with the substrate. Key parameters of the model include the mechanical properties that are measured experimentally using micro-indentation testing. A good agreement between theory and experimental data for both colloidal layers and the real painting is found. These results, by comparing the shrinkage cracks in model layers and in pictorial layers, validate the method based on the use of colloidal systems to simulate and to reproduce drying cracks in paintings. The modeled systems are drying layers of colloidal particles with





Photograph in visible light of the painting "Jeanne d'Arc en prison" by Louis Crignier (1824) – Musée de Picardie, Amiens, © C2RMF/A. Maigret. Inset: Detail of the shoulder showing a drying crack pattern Width of a crack, w, dimensioned by the thickness h of the layer for three different colloidal systems and a pictorial layer. The dashed and plain lines correspond to the model and fit well the experimental results both for colloidal systems and paintings

M. Léang, F. Giorgiutti-Dauphiné, Lay-Theng Lee, L. Pauchard, Crack opening: from colloidal systems to paintings, Soft Matter 34 (2017)

Results achieved within the project DEEPPAINT funded by topic 2 and carried out by Frédérique Giorgiutti-Dauphiné (FAST) and Lay-Thang Lee (LLB).

principles for biofilaments on multiple scales. At the whole cell level, the project has aimed at showing that due to symmetry constraints contractility in disordered networks of filaments and molecular motors proceeds through previously overlooked mechanisms. To understand which of these mechanisms dominates, the genesis of the network's smaller-scale, out-of-equilibrium structure is investigated. Martin Lenz obtained in 2015 an ERC Starting Grant, in the domain of Dissecting active matter.

Key publications

M. Lenz, T. A. Witten, *Geometrical frustration yields fiber formation in self-assembly*, Nature Physics (2017)

G. Foffano, N. Levernier, M. Lenz, *The dynamics of filament assembly define cytoskeletal network morphology*, Nature Communications 7 (2016)

M. Murrell, P.W. Oakes, M. Lenz, M.L. Gardel, *Forcing cells into shape: the mechanics of actomyosin contractility*, Nature Reviews Molecular Cell Biology 16, 486-498 (2015)

The 120k€ of this grant have funded a post-doc student during 2 years. Many collaborations have been initiated, in particular with biologists, and the already existing leverage effects are significant as several local, national and European projects submitted by M. Lenz have been funded.



Examples of frustrated particles and of the fibers they form when aggregated in a computer simulation

A 2nd chair, TURBA (160k \in , 2014-2017) concerns B. Gallet (experimentalist, hired at the end of 2013 in SPEC) and it deals with dissipation in turbulence. The project has involved the realization of several original laboratory experiments that focus on the study of energy dissipation and its "anomalies" in different types of non-isotropic turbulence [Gallet 2017a, Gallet 2017b]. The first setup used is at SPEC and it consisted of a Taylor-Couette device involving drive torque measurements as a function of different rotational speeds of the cylinders. The effect of the anisotropy of the velocity fields was studied as a function of the overall speed of rotation of the fluid. The second experimental setup uses the rotating platform of FAST and it focuses on the same problem, but when the basic flow is the one produced by the rotation of a helix. This project between SPEC and FAST has allowed starting and developing new research actions between the two laboratories. B. Gallet has been able to complement this grant of PALM with a ERC Starting grant.

Key publication

K. Seshasayanan, B. Gallet, Basile; a; Alexakis, *Transition to Turbulent Dynamo Saturation*, Phys Rev Let 119 (2017)

The project of external junior chair NGSTEA of Silvio Franz (75k€, 2013-2016) focuses on the study of the glass transition and glass formation, hampered by the very nature of glassy dynamics itself, which displays an extremely rapid slowing down of the dynamics that inevitably leads to aging when one tries to bring the control parameters close to the values expected for the glass transition [Franz 2016]. Aging makes impossible to collect equilibrium data close to the transition. This is problematic both for measuring critical properties and to test theoretical approaches. Franz has followed two leads to circumvent this difficulty and progress in the study of new kinds of glass transitions (Random Pinning Glass Transition) and in several theoretical, numerical and experimental aspects of aging dynamics able to reveal the physical origin of macroscopic glassy behavior.

Key publication

S. Franz, G. Parisi, M. Sevelev, P. Urbani, F. Zamponi, *Universality* of the SAT-UNSAT (jamming) threshold in non-convex continuous constraint satisfaction problems, SciPost Phys. 2, 019 (2017)

Focus Topic 3 "Ultrafast Dynamics: from radiation sources to multiscale responses"

This topic aims at understanding and controlling the electronic and nuclear motion at the shortest temporal and spatial scales. This implies a direct and intuitive understanding of key fundamental processes, ranging from photoemission and charge migration in molecules, through ultrafast currents and spin dynamics in solids, to coherent collective nonlinear dynamics in plasmas. Over the last decades, considerable investments have been made worldwide to develop tools, fundamental studies and applications of ultrafast dynamics.

The environment of PALM presents an exceptionally rich concentration of advanced light sources and a vast and diverse scientific community active in the field of ultrafast dynamics of matter. The main goal of Topic 3 is bringing together all the researchers working in the field, encouraging the cross-fertilization between novel developments in laser technology and research on the dynamical properties of matter.

The composition of the bureau of PALM-topic 3 reflects this strategy, and its action has been synergetic with the other research structures present in its environment; namely pole 4 of PhOM, which represents a tissue of researchers with a strong overlap with that of PALM-topic 3. This synergy has been possible thanks to a continuous and fruitful exchange among the scientists involved in the coordination of the various structures. In particular, during the last 5 years major infrastructure advances for ultrafast dynamics have been made thanks to the Equipex projects CILEX and ATTOLAB, as well as to the OPT2X program financed by the Idex Paris-Saclay. PALM has played an essential and uniquely complementary role during the construction of these light sources, supporting junior scientists and financing ancillary equipment; the collaborative nature of all PALM projects naturally helps creating a user community around these facilities. Typical examples of this precious action have been two external junior chairs, both of them with a scientific case centered on the possibilities offered by the Equipex projects and both of them resulting in permanent positions: Stefan Haessler, recruited in 2015 and Antonin Borot, recruited in 2017.

The main results obtained in 2015-2017 are reported below. Their presentation is segmented into five sub-topics:

1) Advanced laser and secondary sources, 2) plasmas and high field physics, 3) dilute matter, 4) condensed matter and 5) chemical physics and biological systems.

1. Advanced ultrafast laser and secondary sources

The next challenges for the primary sources for ultrafast dynamics applications are important, diverse and with a strong potential impact. Several major axes appear in these future challenges, such as increasing peak and / or average powers, while increasing repetition rates to extend the application range, realizing shorter pulses in order to reach single or few optical cycle and extending the wavelengths in particular towards the mid-infrared (MIR) range. From a more pragmatic point of view, there is a clear need for greater compactness, efficiency and reliability of laser systems. The need for sources of extremely short pulses, i.e. pulses of the order of the optical cycle, is ubiquitous. This is particularly important for the generation of secondary sources of isolated attosecond pulses. Innovative ultrashort-range amplification techniques, such as OPCPA (optical parametric chirped pulse amplification), are therefore at the center of laser research. For these extremely short pulses, we also see in the current conjuncture a desire to extend these systems to new wavelengths. The realization of optical cycle pulse MIR lasers appears to be very important to allow for the generation of attosecond pulses ever further in the VUV as the cutoff frequency increases with the excitation wavelength. Another extremely important and promising challenge concerns the rise in repetition rates of ultrafast primary sources. This implies an increase in power of the systems with the management of the thermal problems that accompany it. The goal is simple: to increase the recurrence of processes related to the use of pulsed lasers. It also responds to the crucial issue of the creation of high-speed particle sources. In this context, innovations are mainly focused on diode pumped lasers based on ytterbium-doped materials (such as fibre) because they also meet the needs of compactness, efficiency and reliability. <u>PALM financed several projects of source</u> <u>development, often providing seed funding</u> which facilitated the search for matching funds to complete the project, as exemplified here below.

HIGH REPETITION MID INFRARED SOURCES

The majority of high field physics experiments have been performed so far with Ti:Sa laser systems that are able to deliver short (<30 fs) energetic pulses (few mJ) at repetition rate limited to a few kHz, corresponding to average powers below 10 W. However, there is currently considerable technological and research effort towards emerging laser systems that are scalable to higher average powers, and typically operate at much higher repetition rate (>100 kHz) and lower energy per pulse. The OPCPA architecture allows translating the central wavelength to the SWIR (1 μ m - 2.5 μ m) and MIR (> 2.5 μ m) domains. These high repetition rate systems are of great interest for a number of applications such as high harmonic generation (HHG), coincidence detection of photoionization fragments, and photoemission spectroscopy.

The partners of the MIROPCPA project developed such a source, pumped by an ytterbium-doped fiber amplifier, and delivering simultaneously 50 fs, 20 µJ pulses at 1.55 µm and 70 fs, 10 µJ pulses at 3.1 µm at the repetition rate of 125 kHz. This source was used to perform both HHG in solids using the 3.1 µm beam and HHG in gases using the 1.55 µm beam. In Fig. 1, we show the XUV intensity distribution in the spatio-spectral domain and corresponding spectrum obtained in argon. The tight focusing geometry required by the modest pulse energy to perform HHG, together with the long driving wavelength, induce spatio-spectral structures that are currently being investigated.



Figure 1. Spatio-spectral XUV distribution and HHG spectrum obtained in Ar with the MIROPCPA laser

Key publications (project MIROPCPA by Patrick **Georges and Thierry Ruchon)**

A. van de Walle, M. Hanna, F. Guichard, Y. Zaouter, A. Thai, N. Forget, and P. Georges, Spectral and spatial full-bandwidth correlation analysis of bulk-generated supercontinuum in the midinfrared, Opt. Lett. 40, 673-676 (2015)

P. Rigaud, A. Van de Walle, M. Hanna, N. Forget, F. Guichard, Y. Zaouter, K. Guesmi, F. Druon, and P. Georges, Supercontinuumseeded few-cycle mid-infrared OPCPA system, Opt. Express 24, 26494-26502 (2016)

X-RAY LASERS

Besides HHG starting from an optical laser, the other main approach to obtain ultrashort light pulses in the VUV-Xray domain is represented by X-ray lasers, where an optical transition involving core electronic levels is at the heart of the coherent amplification process. There is a strong activity in this field in the PALM community: we present in highlight 9 the results obtained in the framework of the project TEXAS.

See Highlight 9 (project TEXAS by Stephane Sebban)

Highlight 9. Can we shorten the pulse duration of collisional plasma-based soft x-ray lasers below 1 ps?

S. Sebban, A. Depresseux, E. Oliva, J. Gautier, F. Tissandier, A. Lifschitz, Ph. Zeitoun and A. Rousse (LOA), G. Maynard (LPGP)

In plasma-based soft x-ray laser systems, the amplifier lifetime strongly depends on the depletion rate of the lasing-ion population because of collisional ionization during the lasing process. We proposed and demonstrated a novel approach based on the gating of the gain media to reduce the time window in which the lasing action takes place. By increasing the density of the plasma-amplifier, this process becomes fast enough to provoke an anticipated interruption of the amplification process and consequently shortens the duration of the soft XRL pulse [1].



In our work, an ultrashort infrared laser pulse was focused onto an optically preformed waveguiding krypton plasma [2-3] shown on figure a. The resulting electron distribution permits a population inversion by collisional pumping that leads to the lasing of the $3d^94d_{J=0} \rightarrow 3d^94p_{J=1}$ transition of nickel-like krypton at 32.8 nm. When seeded with a high-order harmonic beam, the generated soft X-ray beam exhibits excellent spatial properties [3] as shown on figure b and an adjustable linear polarization [4].

By varying the time delay between the amplifier creation and the seed-pulse injection, we measured the temporal evolution of the amplification factor, and thus revealed the actual temporal profiles of the SXRL gain. In Figure c, the experimental measurements (blue circles) and Maxwell-Bloch-modeling of the amplification factor of the seed pulse (red squares) clearly illustrate the simultaneous temporal quenching of the amplification and the increase of the photon yield. The achieved combination of a shorter duration and a boost in photon yield demonstrates a more than two orders of magnitude upsurge of plasma-based SXRL output power.

A. Depresseux, E. Oliva, J. Gautier, F. Tissandier, G. Lambert, B. Vodungbo, J-P. Goddet, A. Tafzi, J. Nejdl, M. Kozlova, G. Maynard, H. T. Kim, K. Ta Phuoc, A. Rousse, P. Zeitoun, and S. Sebban, Phys. Rev. Lett. 115, 083901 (2015) E. Oliva, A. Depresseux, F. Tissandier, J. Gautier, S. Sebban, G. Maynard, Phys. Rev. A 92, 023848 (2015)

S. Sebban, A. Depresseux, E. Oliva, J. Gautier, F. Tissandier, J. Nejdl, M. Kozlova, G. Maynard, J.P. Goddet, A. Tafzi, A. Lifschitz, H. T. Kim, S. Jacquemot, P. Rousseau, P. Zeitoun and A. Rousse, Plasma Phys. Control. Fusion 60, 014030 (2018)

A. Depresseux, E. Oliva, J. Gautier, F. Tissandier, J. Nejdl, M. Kozlova, G. Maynard, J. P. Goddet, A. Tafzi, A. Lifschitz, H. T. Kim, S. Jacquemot, V. Malka, K. Ta Phuoc, C. Thaury, P. Rousseau, G. Iaquaniello, T. Lefrou, A. Flacco, B. Vodungbo, G. Lambert, A. Rousse, P. Zeitoun & S. Sebban, Nature Photonics 9, 817-821 (2015)

Results achieved in the framework of the project TEXAS funded by topic 3 and carried out by Stephane Sebban (LOA,) and Gilles Maynard (LPGP).

2. High field laser-matter interaction, plasmas

Ultra-High Intensity (UHI) physics investigates the interaction of matter with ultrashort light pulses (5-30fs) at intensities exceeding 10^{18} W/cm². During the interaction, a target of any kind is strongly ionized and a plasma is instantly created. The physics of the fs laser-plasma interaction is very different from that of 'conventional ns plasmas. First, at intensities > 10^{18} W/cm², the motion of electrons in the plasma becomes relativistic. Second, dynamics is dominated by the collective 'coherent' motion of electrons, driven by the (E,B) laser field. The detailed understanding, from elementary laws, of the key processes underlying the apparently complex interaction remains a fundamental challenge. It is also a prerequisite to drive and control collective relativistic motion of matter with laser light.

Such control has many potentially ground-breaking applications. First, ultra-short bursts of coherent light are emitted by the plasma electrons, which spectrally extend up to the X-ray range; and second, beams of high-energy particles (electrons or ions) are expelled from the plasma.

UHI physics can provide table-top sources of high-energy photons and particles which can serve as probes of matter on very small time and space scales. Non-destructive testing or medical imaging and therapy (e.g. cancer protontherapy) are other societal applications.

A key physical system in UHI physics is the plasma mirror (PM), i.e., the dense plasma created at the surface of an initially solid target irradiated by an ultra-intense fs pulse. On fs timescale, the plasma expands over distance << λ_L towards vacuum, so that its interface remains optically-flat. It efficiently reflects the incident UHI beam and can serve as a high-quality mirror to manipulate the beam. An example of application of plasma mirrors will be given below.

At intensity > 10^{18} W/cm², the field can pull *bursts of relativistic electrons* out of PM, once every laser cycle, which emit *light pulses of attosecond duration*, leading to the generation of a train of as pulses, i.e., a spectral comb of *high-order harmonics*. This scheme is one of the best candidates for the next generation of as light sources, to be installed at the Extreme Light Infrastructure (ELI-ALPS, Szeged). Moreover, this scheme even provides the first sources of synchronized ultrashort bursts of light and relativistic electrons, a "dream source" for ultrafast science. Ultrashort bunches of relativistic electrons can also be produced in the interaction of an UHI laser with an underdense plasma. The UHI field excites a large amplitude plasma wave – or wakefield - which can trap and accelerate electrons, providing high quality fs electron beams.

On fundamental side, PM and underdense plasmas provide model systems of a laser field impinging on a well-defined plasma, involving key processes of UHI physics, fieldionization, elastic and inelastic collisions or collective charge motion. Their description constitutes a stringent test for theory. However, while simulations provide "at will" detailed information on the interaction, the observables experimentally accessible are much more limited. Developing a metrology suited to very small temporal and spatial scales of UHI physics is a major challenge.

This is one of the major domains of research covered. We present here some examples of the results obtained over the last three years

ATTOSECOND PULSES IN HIGH FIELD REGIME

The generation of isolated attosecond pulses in the high field regime is one of the major challenges of this field. We present in <u>Highlight 10 the results obtained by one of the junior chairs financed by PALM</u>, Stefan Haessler

See Highlight 10 (Junior chair OPTIWAVE by Stefan Haessler)

LASER WAKEFIELD ELECTRON ACCELERATION

Laser wakefield acceleration capability to sustain fields in excess of 100 GV/m and produce short pulse electron bunches, makes it a promising way towards compact high energy accelerators for a wide range of applications. Multistage acceleration schemes additionally have the potential to provide scalability and control. They are actively investigated for the development of future accelerators, in particular at the Paris-Saclay University within the frame of the CILEX Equipex. In these schemes, an optimized electron injector that produces a high quality electron beam with narrow energy spread and small emittance is one of the key issues.

The partners of the Model-LPA project have investigated experimentally at the UHI100 laser facility and through numerical simulations the injection and acceleration of electrons in the laser wakefield to produce an electron beam that can be efficiently coupled to a plasma accelerator stage. By optimizing the density profile of the gas target a low energy spread can be obtained.

This work is currently being extending, within the ECOLE-ALP project.

Key publications (project MODEL-LPA)

P. Lee, G. Maynard, T. L. Audet, B. Cros, R. Lehe, and J.-L. Vay, *Dynamics of electron injection and acceleration driven by laser wakefield in tailored density profiles*, Physical Review Accelerators and Beams19, 112802 (2016)

B. Cros, B.S. Paradkar, X. Davoine, A. Chancé, F.G. Desforges, S. Dobosz-Dufrénoy, N. Delerue, J. Ju, T.L. Audet, G. Maynard, M. Lobet, L. Gremillet, P. Mora, J. Schwindling, O. Delferrière, C. Bruni, C. Rimbault, T. Vinatier, A. Di Piazza, M. Grech, C. Riconda, F. Amiranoff, *Laser plasma acceleration of electrons with multi-PW laser beams in the frame of CILEX*, Nuclear Inst. and Methods in Physics Research, A, Volume 740, p. 27-33 (2014)

Highlight 10. Shaped laser-wave control of quantum dynamics in atoms and relativistic plasmas.

S. Haessler, R. Lopez-Martens (LOA), M. Turconi, P. Salières, B. Carré (LIDYL)

When high-energy femtosecond laser pulses are focused their electro-magnetic field becomes strong enough to tear electrons off their atoms and subsequently accelerate them along quantum trajectories. Trajectories that recollide with the parent atom may lead to the emission of a train of attosecond (~10⁻¹⁸ s) XUV pulses, two per driving laser optical cycle. Such pulses, generated in gases, are the fundamental tool of the field of attosecond science, studying the movement on bound and quasi-bound electrons in quantum systems. Generating an isolated attosecond pulse requires very short driving laser pulses, not available directly from high energy amplifiers.

Another source of attosecond pulses is created by highly energetic laser pulses, tightly focused onto a solid surface of orders of magnitude higher intensity. These will create a plasma in which electrons are accelerated to nearly the light velocity, and the ensuing relativistic collective dynamics of the plasma electrons lead to the emission of attosecond XUV pulses.

Both sources have in common that the dynamics are driven by the laser field cycles and can therefore be controlled by the shape of these cycles. Standard pulses from femtosecond lasers have a sinusoidal carrier wave. The combination of several laser pulses with different carrier wave frequencies allows shaping the resulting optical field oscillations on the time scale of attoseconds. We have studied, both in theory and experiment, how such cycle-shaped multi-color pulses can be used to control the aforementioned dynamics. For the plasma-based attosecond pulse source, this had never been attempted at the beginning of this project.

For the gas-based attosecond pulses source, we have devised and simulated theoretically a new scheme for limiting the emission of attosecond pulses to a single event per driving laser pulse, which would allow generating an isolated attosecond XUV pulse with very energetic laser drivers of unprecedentedly long pulse duration [1]. The interference of the near infrared laser wave with a suitably tuned longer-wavelength pulse creates a beating that effectively gates the XUV emission. The addition of the second harmonic of the driving laser further enhances the gating and boosts the generation efficiency. The preparation of the experimental realization of this scheme is currently underway at the FAB1 beamline of the EquipEx Attolab, and is expected to convert it to France's first source of isolated attosecond pulses.



For the relativistic-plasma-based attosecond pulse source, we have conducted experiments combing pulses from the UHI laser of CEA Saclay with its second harmonic. The creation of cycle-shaped laser fields at relativistic intensity proved very challenging, but finally suceeded in 2017. By scanning the attosecond phase delay of the two driver-pulse components, we scan the shape of the ultra-intense laser cycles and observe detailed oscillations of both the XUV emission (see figure) as well as the emission of accelerated electrons. While with sinusoidal driver waves, it had always been observed that both types of emission are correlated and maximized in the same conditions, it came as a surprise that the optimal optical- cycle-shape was not found to be the same for the two. Furthermore, we find a higher XUV yield with an optimized two-color driver than with the standard single-color one. Theoretical simulations are now being run that will elucidate the control mechanism for relativistic plasma dynamics behind our experimental observations.

[1] S. Haessler, T. Balcuinas, G. Fan, L. E. Chipperfield and A. Baltuska, *Enhanced multi-colour gating for the generation of high-power isolated attosecond pulses*, Scientific Reports 5, 10084 (2015)

Results achieved in the framework of the project OPTIWAVE funded by topic 3 and carried out by Rodrigo Lopez-Martens (LOA) and Pascal Salières (LIDYL).

SPATIO-TEMPORAL METROLOGY

A new technique of spatio-temporal metrology, named INSIGHT, has been developed by A. Borot (chair junior PALM) and F. Quéré. It enables the measurement of the full intense laser pulse electric field at the focus, which means where the laser-plasma interaction takes place. This has been successfully set-up on UHI100 laser facility, in the experimental room dedicated to electron acceleration. The measurements have led to the identification of spatio-temporal couplings, and in particular a linear variation of the spatial phase frequency before focusing, which presents as, at focus, a wave front direction change with time. This effect is visible on figure 2, which shows the laser pulse reconstruction with space and time, and specifically the

projection on the (x=0,y) plane. Higher order effects have also been observed.



Fig. 2. 3D reconstruction of the E-field amplitude of the UHI100 laser, measured at the focal point, using the INSIGHT technique. On each side, the field projection on each coordinate is reported.

PLASMA MIRRORS AND XUV OPTICAL VORTICES

An example of applications of plasma mirrors to manipulate high field radiation in the XUV domain is presented in Highlight 11. The production and control of light carrying orbital angular momentum is a major challenge, in particular at short wavelengths. This is an example of how <u>PALM</u> <u>finances equipment to support experiments performed on</u> the large facilities of its campus.

See Highlight 11 (Projects PTYCHOGRAT and PLASMOAM by Fabien Quéré)

3. Dilute matter

Gas phase UFD considers as-fs-ps electronic/nuclear processes in isolated systems, i.e., atoms, molecules, clusters and nanoparticles, or molecules on solid surface. It provides useful models of systems strongly coupled internally or with a light field, and weakly coupled with their environment, e.g., biomolecules interacting with natural light. Though "natural" dynamics triggered by incoherent excitation is statistically incoherent, its microscopic quantum mechanical essence is coherent, especially at short time and is fruitfully approached by "artificial" wavepacket dynamics allowing systematic studies of, e.g., photoionization at the

attosecond timescale, charge migration, or the very first steps of the coupling between electronic and nuclear motions. By increasing the system complexity, e.g., in large molecules and clusters, one can investigate the transition from isolated coherent to collective dynamics and decoherence.

Theoretically, isolated systems provide the ideal frame for describing basic ultrafast processes. However, even in simple cases (e.g., He atom in strong field), this remains a challenge.

Ultrafast processes are usually triggered by light, in either the weak-field perturbative regime (intensity $<10^{12}$ W/cm²), or the strong-field non-perturbative regime (intensity $\geq 10^{12 \cdot 16}$ W/cm²). In the weak-field regime, since atto pulses have low energy (10-100 nJ in ~10 eV BW), only a few XUV/XUV studies have been reported yet. Most studies combine as-fs XUV with UV-vis-IR laser pulses of which the one-cycle electric field provides the synchronized time gradient, sometimes at the limit of perturbative conditions. Production of intense atto pulses, either laser- or FEL-driven, is a technological challenge of UFD. In parallel, the development of theoretical tools and experimental techniques, to study small/large systems in perturbative/non-perturbative interaction, opens long term perspectives to gas phase UFD.

Highlight 11. Generation of optical and XUV vortices at ultra-high Intensity

A. Leblanc, A. Denoeud, L. Chopineau, G. Mennerat, Ph. Martin and F. Quéré, (LIDYL)

Optical vortices are light beams with helical wavefront carrying orbital angular momentum (OAM). Due to their amazing properties (spiral phase and ring intensity), these beams have already paved the way for new applications (spectroscopy, microscopy, optical manipulation ...) in the visible and infrared domains, which one seeks to extend in the X-UV domain. Today, these optical vortices are generating increasing interest in the ultra-intense laser community, as they offer new ways to control interactions with matter and generate XUV and ultra-short particle sources with original properties. Although a large number of theoretical studies have been carried out on the subject, no experiment has yet demonstrated these effects for very high intensities (I> 10¹⁶ W / cm²), given the difficulty of producing a large and intense laser beam with a helical wavefront. We have just shown, on the UH1100 laser laser facility, that it is possible to generate such pulses carrying OAM and to transfer this moment to a beam of harmonics generated on a plasma mirror for intensities higher than 10¹⁹ W / cm². For this, we introduced a helical phase plate in the path of a high-intensity infrared laser beam, to produce vorticity, and studied the X-UV vortices generated by interaction with a glass slide. At the same time, we have developed a method based on holography, to provide OAM to the most intense laser beams (see figure). This technique is particularly interesting for handling and shaping any type of ultra-intense laser beam, since it has the enormous advantage of being able to sustain considerable intensities without requiring any large optics which are generally demanded for this kind of beam.



An initially flat solid target is ionized by an intense holographic pattern (peak fluence of $\approx 103 \text{ J cm}^{-2}$ in bright fringes), obtained by crossing a reference and an object beam (in blue), both consisting of femtosecond pulses. In this example, the holographic pattern is a fork grating (white and grey image in the target plane), produced by placing a spiral phase plate in the object beam. b, The spatial modulation of the ionizing beam fluence leads to a modulation of the plasma expansion velocity. This results in the growth of a structured plasma surface in the picoseconds following ionization—in the present example, a plasma fork grating. c, This structured plasma surface can diffract an ultraintense femtosecond laser beam, leading to high-energy replicas of the initial object beam in the diffracted orders

A. Leblanc, A. Denoeud, L. Chopineau, G. Mennerat, Ph. Martin, F. Quéré, *Plasma holograms for ultrahigh-intensity optics*, Nature Physics 13, 440 (2017)

Results achieved in the framework of the projects PLASMOAM and PTYCHOGRAT funded by topic 3 and carried out by Fabien Quéré (LIDYL),

We present here selected examples of the <u>support provided</u> by PALM to this field, in particular by financing young <u>scientists</u> working on these projects.

ATTOPHYSICS

XUV/XUV studies would be greatly facilitated by the generation of perfectly synchronized but spatially separated atto-pulses. The attosecond lighthouse effect was proposed and first demonstrated by PALM researchers in 2012. Recent studies include theoretical investigations of the impact of the spatio-temporal structure of the driving laser field (see Fig. 3) on phase matching of the high harmonics emitted in gases.

Key publication (project IMAPS)

T. Auguste, O. Gobert, T. Ruchon, and F. Quéré, *Attosecond lighthouses in gases: A theoretical and numerical study*, Physical Review A 93, 033825 (2016)

Determining the dynamics of the spectral amplitude as well as of the phase of photoelectron wavepackets with attosecond resolution is one of the current challenges of attophysics: recent experiments supported by PALM are presented in Highlight 12.

See Highlight 12 (project IMAPS by Thierry Ruchon)



Figure 3: Space-time electric-field distribution calculated at focus of the driving laser (a), and at 0.5x (b), 1x (c) and 2.5x (d) the Rayleigh length, in the case of the attosecond lighthouse effect where a wavefront rotation is induced on the beam.

Highlight 12. Attosecond spectroscopy of resonant photoionization dynamics

V. Gruson, L. Bareau, T. Ruchon, A. Borot, B. Carré, P. Salières (LIDYL), F. Martin et al (UAM-Madrid) and R. Taieb et al (LCMPR-Paris)

Spectroscopy has taught us how the very precise measurement of resonance lineshapes gives insight into the structure of matter. However, as a time-integrated measurement, the spectral lines give only indirect information on the underlying electronic dynamics. The resonance width can be related to the timescale of the electronic excitation and relaxation, but, in the general case, this is not enough for accessing the details of the full dynamics that have to be recovered from advanced modeling. A typical case is the one of autoionizing resonances, where the system (atom, molecule, nanostructure) can be ionized either directly to the continuum or be trapped in a very excited state for a very short time (femtosecond) before reaching the continuum. The interference between the two channels results in an asymmetric lineshape, called Fano profile after the Italian theoretician Ugo Fano who first modeled this process. While the Fano profile has been extremely successful in analyzing the absorption lines measured in a wide variety of systems, the details on how the process unwraps in time have remained elusive, the ultrashort timescale at stake precluding direct time-domain investigations.

We have developd a new technique, based on spectrally resolved electron interferometry in combination with ultrashort XUV and IR pulses, allowing the measurement of the spectral amplitude and phase of the resonant photoelectron wave packet. In this scheme, replicas obtained by perturbative two-photon XUV+IR transitions interfere with reference wave packets that are formed through smooth continua, allowing the full temporal reconstruction, purely from experimental data, of the resonant wave packet released in the continuum. In turn, this allows resolving the ultrafast buildup of the autoionizing resonance, revealing the decomposition of the process in two nearly consecutive steps governed by fairly different time scales: during the first 3 fs, the direct ionization channel dominates; then, the resonant path starts contributing as the doubly excited state decays in the continuum, resulting in interferences between the two channels that ultimately shape the celebrated Fano profile [1]. This opens multiple opportunities for studying ultrafast strongly correlated dynamics in a variety of systems, from molecules and nanostructures to surfaces, and controlling matter changes at a most fundamental level.



Reconstruction of the ultrafast buildup of the Fano 2s2p resonance of helium

V. Gruson, L. Barreau, À. Jiménez-Galan, F. Risoud, J. Caillat, A. Maquet, B. Carré, F. Lepetit, J-F. Hergott, T. Ruchon, L. Argenti, R. Taïeb, F. Martin and P. Salières, *Attosecond dynamics through a Fano resonance: monitoring the birth of a photoelectron, Science* 354, 754 (2016)

Results achieved in the framework of the project IMAPS funded by topic 3 and carried out by Thierry Ruchon (LIDYL), Rodrigo Lopez-Martens (LOA) and Franck Delmotte (LCF).

ATTOSECOND MOLECULAR SPECTROSCOPY

The achievement of time-resolved images of the coupled electronic and nuclear dynamics in molecules at the atto-tofemtosecond time scale is a hot topic in molecular physics. Photoionization by XUV pulses is currently involved in such studies either to initiate electronic dynamics in the continuum or to probe the relaxation of transient excited molecular states. Taking advantage of dissociative photoionization, the ImDynCo project combines the use of electron-ion coincidence momentum spectroscopy and that of high order harmonic generation on the 10 kHz beamline at ATTOLAB to access most complete observables consisting of timeresolved molecular frame photoelectron angular distributions (MFPADs) [Vevrinas 2016]. with complementary spectrally resolved experiments at SOLEIL.

For extended studies on the role of electron and nuclear Degrees Of Freedom (DOFs) in photoionization in the nonlinear regime, higher pulse intensities are necessary. Free electron lasers constitute such powerful sources of femtosecond XUV pulses and the FERMI facility is unique for the production of coherent VUV pulses.

First studies of non-linear two-photon dissociative (DI) and non-dissociative (NDI) photoionization of H₂ and D₂ involving resonant excitation of vibrationally resolved intermediate molecular excited states have been performed at the FERMI Low Density Matter beamline. They demonstrated that this process leads to a remarkable enhancement of the DI/NDI ratio by about two orders of magnitude relative to the onephoton ionization at comparable excitation energy. Furthermore, the DI/NDI ratio increases with increasing vibrational levels, highlighting the control of the photoionization outcome by the nuclei DOF of the hydrogenic molecule. This trend supports the striking predictions of Time Dependent Schrödinger Equation ab initio calculations. Velocity Map Images (VMI) of the emitted photoelectrons and photoions were recorded, this technique being well adapted to the low 10-50 Hz repetition rate of FERMI. These energy and angularly resolved results elucidate the main reaction pathways of DI and NDI and establish unambiguously the enhanced role of repulsive excited states of the H₂⁺ molecular ions in DI.

Key publications (project ImDynCo by Danielle Dowek)

K. Veyrinas, V. Gruson, S.J. Weber, L. Barreau, T. Ruchon, J.-F Hergott, J.-C. Houver, R.R. Lucchese, P. Salières, and D. Dowek, *Molecular frame photoemission by a comb of elliptical high-order harmonics: a sensitive probe of both photodynamics and harmonic complete polarization state*; Faraday Discussions, 194, 161 (2016)

INNER SHELL SPECTROSCOPY

The excitation of core electronic levels at shorter wavelengths, in the X-ray domain, is another important component of the gas phase activity of PALM. We present here results obtained in the framework of the Senior Chair for F. Gelmukhanov.

See highlight 13 (project Ultrafast-X by Catalin Miron)

4. Condensed matter

The study of out-of-equilibrium dynamics in solids has become a major topic of modern condensed matter physics. Out-of-equilibrium states can be obtained by irradiation with ultra-short laser pulses, which induce electronic transitions on a timescale at which the lattice can be regarded as frozen. One can therefore transiently decouple the different degrees of freedom of the system (electronic, lattice, spin, orbital), giving the possibility of learning about their mutual interaction during the recombination process, and in certain cases of modifying this interaction in a controlled way.

The activity in the PALM community covers several research lines (ultrafast magnetization [Schmising 2014], coherent lattice vibrations [Esposito 2015], Dirac fermion dynamics in topological insulators [Khalil 2017], out of equilibrium strongly correlated materials [Jacques 2016], plasmonics, HHG in solids, semiconductors, 2D materials). PALM contributed to this activity by *financing young scientists as well as the development of equipment of mutual interest* among collaborating laboratories. We present here some of the results obtained in the field.



Figure 4. Two photon photoionisation in molecules

Highlight 13. Einstein-Bohr gedanken experiment performed at the molecular level Catalin Miron (SOLEIL), Bertrand Carré (LIDYL) et Osman Atabek (ISMO)

For the first time, an almost centenary thought experiment proposed by Albert Einstein and Niels Bohr in their discussion on whether the elementary particles constituting our surrounding world should be seen as particles or waves, was successfully realized at the molecular level (O2), by synchrotron based experiments. This result is adding a new cornerstone to one of the richest public debates in the history of Science, triggered by the original recoiling double-slit gedanken experiment, which has already led to several Nobel Prizes. We showcase a materialization of this gedanken experiment by resonant X-ray photoemission from molecular oxygen for geometries near equilibrium (coupled slits), and in a dissociative state far away from equilibrium (decoupled slits). Interference is observed in the former case, while the electron momentum transfer quenches the interference in the latter case owing to the Doppler labelling of the counter-propagating atomic slits, in full agreement with Bohr's complementarity.



Schematic representation of the double slit gedanken experiment, with coupled (a) and decoupled (c) massive slits, and the schematics of the materialization of this thought experiment using a molecular photoionization experiment where the two slits are replaced by two coupled (b) and decoupled (d) oxygen atoms. The recoil momenta and the propagation directions of the dissociating atoms and electron are shown schematically by yellow, green and dashed blue arrows, respectively

X-J. Liu, Q. Miao, F. Gel'mukhanov, M. Patanen, O. Travnikova, C. Nicolas, H. Ågren, K. Ueda & C. Miron, *Einstein–Bohr recoiling double-slit gedanken experiment performed at the molecular level, Nature Photonics*, 9 (2) 120-125 (2015)

Results achieved in the framework of the project ULTRAFAST-X funded by topic 3 and carried out by Catalin Miron (SOLEIL), Bertrand Carré (LIDYL) and Osman Atabek (ISMO).

OUT OF EQUILIBRIUM CORRELATED MATERIALS

The phase diagrams of strongly correlated materials reflect the complex interplay between electrons, phonons, spins and orbital degrees of freedom, and offer exciting opportunity of ultrafast studies. Experimentally, the effects of ultrafast photoexcitation take various remarkable forms, such as photoinduced phase transitions, quasi-particle dynamics in superconductors, ultra-fast symmetry changes and the occurrence of new, transient phases of matter. From a theoretical perspective, the nontrivial and rich transient dynamics of correlated electron systems has triggered novel investigations, questioning, e.g., the thermalization of pumpexcited Mott or Kondo insulators and the role of lattice vibrations, orbital degrees of freedom and competing orders in the relaxation dynamics. Materials where the effects of strong electron correlations are intertwined with those of strong spin-orbit coupling, like for instance iridates, present very interesting opportunities for their ultrafast manipulation. An exemple of the activity of PALM in this domain is the study of materials presenting metal-insulator transitions, like the prototype Mott-Hubbard compound vanadium sesquioxide, V₂O₃. The experiments performed on out-ofequilibrium V₂O₃ have shown a surprising hardening of the ion lattice while the electrons are still in an excited state, revealing a novel state of matter whose lifetime is only of few picoseconds (Fig. 5). This transient state plays a crucial role in the relaxation process following photoexcitation, and cannot be achieved by modifying conventional thermodynamic parameters such as temperature or pressure.

Key publication (project FastMap by Marino Marsi)

G. Lantz, B. Mansart, D. Grieger, D. Boschetto, N. Nilforoushan, E. Papalazarou, N. Moisan, L. Perfetti, V. L. R. Jacques, D. Le Bolloch, C. Laulhe, S. Ravy, J.-P. Rueff, T.E. Glover, M.P. Hertlein, Z. Hussain, S. Song, M. Chollet, M. Fabrizio & M. Marsi, Ultrafast evolution and transient phases of a prototype out-of-equilibrium Mott-Hubbard material, Nature Communications 8, 13917 (2017)



antibreathing mode, as sketched on the right), corresponding to a non thermal transient behaviour for both the metallic and the insulating phase
HHG IN SOLIDS/PLASMONICS

Non-linear optical processes such as high harmonic generation (HHG) rely on strong light-matter interaction. This can be achieved by squeezing light in space, compressing light in time, or by both. Traditionally HHG occurs in rare-gas atoms at an intensity of about 1014W/cm² achieved by means of mJ-level amplified lasers. This clearly prevents the development of hand-held systems. In 2011. Stanford and Ohio State Universities put in evidence for the first time HHG from a bulk semiconductor crystal. This experiment started a vast effort by numerous laboratories towards the understanding of the phenomenon, proposing different models and pushing experimental setups. We have recently found numerically a new effect so-called "near field phase matching" for the control of attosecond pulses using plasmonic field enhancement. Following this, recent experimental findings at CEA report on the third harmonic generation (THG) from two original nanostructures in the sub-terawatt regime. We first investigate a bi-laver structure composed of a thin ZnO crystal and an out-coupling gold layer with a 2D array of nanoholes showing extraordinary transmission. The THG signal is strongly amplified and shows polarization independent field amplification. To increase the light coupling and polarization selectivity we then introduce a novel hybrid Metal-Dielectric-Metal (MDM) metasurface that induces a boosted amplification of the THG compared to the bi-layer structure (see Fig. 6). Indeed, the Fabry-Perot plasmonic resonator allows a high confinement and amplification of the laser electric field over a large volume of the SiO2 dielectric film. THG signal is amplified by one order of magnitude. The THG polarization dependence with respect to the fundamental laser allows an ON/OFF switch of the enhancement and confirms the plasmonic origin of the amplification (see polarisation dependence in Fig. 6). However these experiments also suggest damage threshold and lifetime limitations of plasmonic metallic nanostructures at IR wavelength. To go beyond, we have designed an all-semiconductor field nano-amplifier based on an array of ZnO nanocones with resonance in the midinfrared. The laser intensity is enhanced locally by more than one order of magnitude as supported by FDTD simulation. Strong amplification of the 7th harmonic from our all 3D waveguide are reported. Lifetime of such structure exceeds a week which opens applications of this novel nano-source of hot photons.



Key publications (projects HILAC by Willem Boutu and PLASMON-X by Hamed Merdji)

T. Shaaran, R. Nicolas, B. Iwan, M. Kovacev, H. Merdji, Nanoplasmonic near field phase matching of attosecond pulses, Scientific Reports 7, 6356 (2017)

L. Shi, B. Iwan, R. Nicolas, Self-optimization of plasmonic nanoantennas in strong femtosecond fields, Optica 4, 1038 (2017)

5. Chemical and biological systems

Ultrafast dynamics is an interdisciplinary field, and many interesting scientific programs are at the border between physics and other domains, like chemistry or biochemistry. We present here two examples.

CONFORMATION-SELECTIVE DYNAMICS OF COMPLEX SYSTEMS

Spectroscopy coupled to dynamics studies is a powerful way to investigate structural effects in complex systems. For example, the absorption spectrum of the deposited DABCO molecule on a large argon cluster (about 500 atoms) shows two solvation sites populated in the ground state. The combined dynamics study shows that the population ratio of the two sites is reversed when the molecule is electronically excited, providing the timescale (0–6 ps) of the corresponding solvation dynamics.

The DIRCOS project aimed at applying such a strategy to study how UV absorption by flexible biomolecules, such as proteins or peptides, significantly modifies the interactions at play within these molecules and therefore their structure. IR spectroscopy is a tool of choice for characterizing H-bonds within a biomolecule, especially because these bonds control their shape and their structure. The joint use of UV and IR lasers even makes it possible to record conformationselective IR spectroscopy, providing an elegant access to the structure of each conformation. The IR / UV spectroscopic technique, commonly used to document the structure of jet-cooled molecules in their ground state, has been extended to excited states, whose IR spectrum is obtained by a UV / IR / UV laser excitation scheme coupled with an ion detection by mass spectrometry. The comparison of the IR spectra of the ground state and the electronic state makes it possible to assess the modification of the structure of each conformation upon electronic excitation. The red shift of the frequency of the elongation vibration of an NH bond in a model peptide (Figure 7) upon excitation will indicate an enhancement of the interaction in the excited state. These precise spectroscopic measurements provide benchmark data for evaluating performances of the quantum chemistry methods used to model these systems, notably in the excited state.

The use of an intense trapping laser for the levitation could lead to an increase of the internal temperature of the levitated nanoparticle. The particle heating will then limit the stability of the system and will impact the centre of mass motion of the particle. This effect could mask quantum effects and will limit force sensitivity of the apparatus. The measure and the control of the internal temperature of the levitated nanoparticle are then crucial for the development of levitation experiments. The levitation of a nanodiamond hosting NV defects, which constitute a sensitive thermometer, is an essential step toward the understanding of the impact of internal temperature on the particle dynamics. It should also provide a solution for a better control of the particle heating.



Figure 7. (Top) Structure of the $\gamma(g+)$ conformation of the phenylalanine dipeptide, stabilized by 7 and π H-bonds. (Bottom) The IR spectra of the ground (S0) and first excited (S1) states obtained by IR/UV and UV/IR/UV photonisation schemes. The highest frequency of the IR spectrum corresponds to the elongation motion of the NH bond involved in a π H-bond with the π electron density of the phenyl ring. Its red shift upon UV excitation (red line) indicates a stronger interaction in the S1 state.

Key publication (project DIRCOS by Lionel Poisson)

WY Sohn et al., Local NH- π interactions involving aromatic residues of proteins: influence of backbone conformation and $\pi\pi^*$ excitation on the π H-bond strength, as revealed from studies of isolated model peptides, Phys.Chem.Chem.Phys., 18, 29969 (2016)

FIRST PRINCIPLES SIMULATION OF ATTOSECOND ELECTRON DYNAMICS IN COMPLEX SYSTEMS

Recent years have seen a growing interest in the electron dynamics taking place in molecules when they are subject to an external perturbation. This interest has been stimulated by progress in attosecond spectroscopy that now gives access to details on electron dynamics. The realm of subfemtosecond electron dynamics involves fascinating processes such as ultrafast charge migration, Auger decays and Intra Coulomb Decays. The photophysics of molecules of biological interest like metalloporphyrins is also a hot topics where ultrafast electron dynamics comes into play. Facing the remarkable progresses in experiments, there is a huge need to develop first principles approaches to simulate these ultrasfast processess at the microscopic level.

In the PHOTOHEME project a unique computational set-up based on Real-Time Time-Dependent Density Functional Theory (RT-TDDFT) has been devised. It allows simulations of the irradiation of condensed matter either by electric fields (of moderate to intense intensities) or by high energy charged particles. The Figure 8 shows autocorrelation function of the average dipole moments of each hydration layer of a peptide subject to a short electromagnetic pulse. The curves hihlight the speed at which the energy initially deposited in the peptide is dissipated into the environment (damped oscillations). We are now now ready to apply this methodology to study the non-adiabatic dexcitation decay of metalloporphyrins. The realization of this project funded by PALM is also finding many other applications at LCP, notably on the modelling of radiation induced damages in biology by first principles approaches.

Key publications (project PHOTOHEME by Aurélien de la Lande)

A. de la Lande, M. Ha-Thi, S. Chen, B. Soep and N. Shafizadeh, Structure of cobalt protoporphyrin chloride and its dimer, observation and DFT modeling, Phys. Chem. Chem. Phys. 18, 16700-16708 (2016)

X. Wu, J. -M. Teuler, F. Cailliez, C. Clavaguéra, D.R. Salahub and A. de la Lande, Simulating Electron Dynamics in Polarizable Environments, J. Chem. Theor. Comput. 13, 3985- 4002 (2017)



Figure 8 Ultrafast evolution of the autocorrelation function of an out-of-equilibrium peptide

Topic 4 "Emergence, Evolution, Rapid Reaction"

This topic is devoted to promote the opening of new collaborations that arise outside the field of the three PALM focus topics. The 38

board has particularly supported projects where the sharing of knowledge between teams was particularly strong and which opened the opportunity of a better scientific structuration of the LabEx PALM community. In this spirit the biological soft matter people were identified as a community strong enough to be incorporated in the "Slow and Emergent Dynamics in Out-of-Equilibrium Systems" focus topic. We have funded projects in the fields of (i) new materials, (ii) biosciences, (iii) plasma chemistry, (iv) free radicals and molecules spectroscopy (v) Advanced instrumentation and (vi) exploratory projects. These projects are related to advanced chemical physics and physical chemistry, including elementary biological studies. One main line is the study and control of energy transfer processes; another main line is the fine analysis of the properties of matter in order to get precise information on the values of the parameters that characterized it. The projects that have been submitted have covered mostly the interfaces between the LabEx PALM teams. An increasing part of them is concerned by interfaces involving research teams outside the LabEx PALM community.

1. New Materials

ECRIN (collaboration between NIMBE, SOLEIL and LCP) focusses on Imogolite, a natural nanotubular aluminum silicate clay mineral, originally found in volcanic soils. Its versatile structure is well adapted for the study of water confined in a one-dimensional structure. Two types of synthetic imogolites whose outer surface is covered with Al-OH groups were investigated: a pristine imogolite (IMO-OH) with a hydrophilic inner surface fully covered with Si-OH groups and a hybrid imogolite (IMO-CH3) with a hydrophobic inner surface fully covered with Si-OH groups. Insights into the relationship between (1) the large specific surface area, (2) restricted geometries, (3) the tunable hydrophilicity, can serve the scientific community interested in deploying these materials for applications such as membranes, catalyst supports, adsorbents, etc.

See Highlight 14 (project ECRIN by Sophie Le Caer)

REMI (collaboration between LSI and ICCMO) investigates the possibility to control the Eu³⁺ environment in different metaphosphate and polyphosphate glass compositions under irradiation. The study focuses on the mechanism leading to the evolution of the glass by comparing the impact of high energy electron and fs laser. Using SIRIUS Accelerator, glasses were irradiated with electrons of 2.5 MeV at various doses. There is no important change of the metaphosphate glass structure whatever the integrated dose contrary to polyphosphate glasses that show significant change (glass depolymerization) of the vitreous network from 108Gy. The evolution of the Eu³⁺ ion environment is studied by photoluminescence. The asymmetry ratio between Forced electric dipole and Magnetic dipole is used to characterize the deviation and the distortion of the site symmetry. The irradiation dose leads to important increase of the asymmetry ratio an in metaphosphate glasses and to a moderate one in polyphosphate glasses. It shows that the local site symmetry of the ion is disconnected to the network variation even if in both types of glasses an additional reduction of Eu³⁺ into Eu²⁺ under irradiation is observed.

ANTIDOX (collaboration between SPEC, SOLEIL, UMPhy) studies how spin currents can produce a magnetic torque on the antiferromagnetic order in order to get active elements in spintronics. This opens the fields of spintronics and magnonics to antiferromagnetic (AF) insulators. The goal is to demonstrate that pure spin currents, strain and/or femtosecond light pulses can be used to manipulate the AF order in some well-chosen AF insulators. Several external stimuli have been tested to attempt writing AF domains, in order to use insulating antiferromagnets as memory elements. The technique is compatible with time-resolved imaging using pump-probe methods. The time evolution of the AF domains is thus achievable at a time-scale of the order of 100 fs. This allows probe antiferromagnetic dynamics, which is expected in the THz range.

See Highlight 15 (project ANTIDOX by Michel Viret)

FEMTONIC³ (collaboration between LSI and TRT) is devoted to the study of electron scattering by phonons one of the major processes that determine the transport characteristics and relaxation dynamics in semiconductorbased devices. In photovoltaic cell, the solar energy absorbed by electrons and exceeding that of the band gap is rapidly lost due to interactions with crystal vibrations (phonons). Detailed understanding of the hot carrier relaxation processes is thus necessary in order to make possible the device optimisation and the search for new materials. A new computational method, based on the interpolation of the electron-phonon matrix elements in Wannier representation, allows the calculation of the electron-phonon coupling in polar materials. This allows interpret the dynamics of hot electron relaxation in bulk GaAs. The electron-phonon scattering times turned out to be surprisingly fast, of the order of a few tens of femtoseconds. The relaxation of hot carriers in GaAs, one can find two distinct relaxation regimes, one related with the momentum, and the other with energy relaxation. Both regimes are almost entirely ruled by the electron-phonon interaction.

³J. Sjakste, N. Vast, M. Calandra, F. Mauri, *Wannier interpolation of the electron-phonon matrix elements in polar semiconductors: Polar-optical coupling in GaAs, Physical Review B 92, 054307 (2015). H. Tanimura, J. Kanasaki, K. Tanimura, J. Sjakste, N. Vast, M. Calandra, F. Mauri, <i>Formation of hot-electron ensembles quasi-*

equilibrated in momentum space by ultrafast momentum scattering of highly excited hot electrons photoinjected into the Γ valley of GaAs, Physical Review B 93, 161203 (R) (2016)

Highlight 14. Properties of Water Confined in Standard and Hybrid Imogolites Revealed by Infrared Spectroscopy

Yuan-Yuan Liao, Pierre Picot, Antoine Thill, Sophie Le Caër (LIONS), Jean-Blaise Brubach, Pascale Roy (Synchrotron SOLEIL)

Imogolite is a natural nanotubular aluminum silicate clay mineral, originally found in volcanic soils. Its well-defined, yet tunable structure makes it a good candidate for the study of water confined in a one-dimensional structure. Two types of synthetic imogolites were investigated: a pristine imogolite (IMO-OH) with a hydrophilic inner surface fully covered with Si-OH groups and a hybrid imogolite (IMO-CH₃) with a hydrophobic inner surface fully covered with Si-OH groups. The structures are presented in Figure (a).

We succeeded in producing self-sustaining thin films of imogolites. The behavior of water confined in these films was then studied by infrared spectroscopy in the 20-4000 cm⁻¹ spectral range as a function of relative humidity (RH, %). The water adsorption properties were determined and compared in details for both imogolites. By using the typical external water spectrum deduced for IMO-CH3, we have successfully extracted the spectrum of water confined within IMO-OH nanotubes. Figure (b) shows then the water adsorption isotherm of water confined inside IMO-OH and the corresponding IR spectra obtained during the water-adsorption process.

The water filling inside IMO-OH follows these steps: I) Water molecules, adsorbed to silanol groups on the internal surface, start to form patches. II) Those water patches merge into a monolayer over the entire internal surface. III) Those patches grow and then form plugs. IV) Pores are filled with water.

Lastly, insights into the relationship between (1) the large specific surface area, (2) restricted geometries, (3) the tunable hydrophilicity, can serve the scientific community interested in deploying these materials for applications such as membranes, catalyst supports, adsorbents, etc.



Y-Y. Liao; P. Picot; J.-B. Brubach; P. Roy; S. Le Caër, A. Thill, Self-supporting thin films of imogolite and inogolite-like nanotubes for infrared spectroscopy, Applied Clay Science (2017)

Y-Y. Liao; P. Picot; M. Lainé; J.-B. Brubach; P. Roy; A. Thill; S. Le Caër, *Tuning the Properties of Confined Water in Standard and Hybrid Nanotubes: an Infrared Spectroscopic Study*, submitted (2017)

Results achieved in the framework of the project ECRIN funded by topic emergence and carried out by Sophie Le Caer (NIMBE), Pascale Roy (SOLEIL) ansd Mehran Mostafavi (LCP).

ASMOE (collaboration between ISMO and SOLEIL) investigates how ultrathin alkali-halide films react upon electron irradiation. These films are increasingly used as spacers to electronically decoupled organic molecules from metallic substrates in atomically controlled systems for single molecule spectroscopy measurements. Their modification upon electron irradiation had to be addressed. In the case of bulk alkali-halide crystals, electron irradiation is known to induce layer-by-layer alkali halide desorption, through a mechanism that involves the creation of halogen atom vacancies in the bulk. In the ultrathin film case, the limited amount of reactants and the interactions with the substrate are crucial, leading to different reaction kinetics, compared to bulk crystals. The outcomes may be different too, since the products of alkali-halide dissociation may adsorb or even react with the substrate, whereas on bulk crystals the dissociation products inevitably adsorb on the same alkali-halide material.



The desorption of ultrathin NaCl films grown on Ag(001) upon electron irradiation is studied using LEED, AES and STM. LEED and AES were used both to modify the NaCl films and to investigate the reaction kinetics and products. Na atoms produced from NaCl dissociation diffuse to bare areas of the Ag(001) surface, where they form Na-Ag 40

superstructures that are known for the Na/Ag(001) system. The electron irradiation induces an increasing disorder in the ultrathin NaCl films, which slows down the reaction kinetics; as a result, the reaction kinetics and products depend on the NaCl film thickness. This study is a first step toward the use of electron-irradiated ultrathin alkali-halide films as "templates" for anchoring single organic molecules or producing macromolecular structures on a surface.

Key publication

A. Husseen, S. Le Moal, H. Oughaddou, G. Dujardin, A. Mayne, E. Le Moal, *Reaction kinetics of ultrathin NaCl films on Ag(001) upon electron irradiation*, Physical Review B (2017)

EXIMOL (collaboration between LPS and LAC) studies magnetic field effect in Silicon light emitting diodes. Spin dependent magnetic field effects on electro/photoluminescence are highly specific to organic semiconductors. The presence of similar physics in silicon light emitting diodes (SiLEDs) has been investigated. SiLEDs are a-priori very inefficient due to the indirect band-gap in Si and sophisticated device geometries are usually needed to improve their efficiency. This precludes magnetic field effect studies as it is then impossible to separate classical magneto-resistance from spin processes. This challenge was achieved fabricating SiLEDs using gas immersion laser doping which allowed us to achieve efficient emission while retaining a well defined planar geometry where the alignement between electric and magnetic fields suppresses classical magnetoresistance to within a few percent. In this

Highlight 15. Imaging Antiferromagnets using Second Harmonic Generation M. Viret, J.-Y. Chauleau, T. Chirac (SPEC), S. Fusil and C. Carétéro (UMPhy)

Spintronic devices are largely based on ferromagnetic metals (FM) in multilayer stacks. So far, antiferromagnets (AF) are only useful indirectly, because of their coupling which pins the magnetic moments of FM layers. The realization that spin currents can produce a magnetic torque on the AF order opens a new route towards introducing these materials as active elements in spintronics.

In CEA/SPEC, we have started a new activity aiming at opening the fields of spintronics and magnonics to antiferromagnetic (AF) insulators. Our goal is to demonstrate that pure spin currents, strain and/or femtosecond light pulses can be used to manipulate the AF order in some well-chosen AF insulators. Imaging techniques are therefore essential because one needs to visualize AF domains to assess the various writing parameters. Because of the absence of stray fields in these materials, imaging is a very challenging task. Presently, the most efficient techniques rely on using linearly polarized light (synchrotron or visible) to measure the direction of the AF vector. However, another technique based on non-linear optics can also be very powerful. Indeed, when illuminated by intense light, some non-linear materials can re-emit higher harmonics. This is allowed in non-centrosymmetric materials as well as on



surface and interface states of centrosymmetric media owing to the breaking of inversion symmetry at the boundaries. Similarly, the breaking of time-inversion symmetry by long-range magnetic ordering leads to new contributions to SHG. As these depend on the direction of AF vectors, they can be used to probe the magnetic structure.

We have recently setup such a high-resolution SHG imaging in transmission mode in our laboratory. It uses laser pulses emitted from an amplified Ti-Sapphire laser whose wavelength can be continuously adjusted by Optical Parametric Amplification from 2600 nm to 530 nm. The emitted light impinging on the sample is linearly polarized and can be rotated using a λ /2 prism. Right after it, a custom-made interference filter completely rejects the first harmonic light, before the second harmonic is collected by a high numerical aperture objective and is imaged by a sensitive Peltier cooled CCD camera (acquired through the LabEx).

Typical antiferromagnets like NiO have typically 12 kinds of AF domains, which makes any attempt to disentangle magnetic contrasts a difficult challenge. This situation can be largely simplified in multiferroics, materials where another order can couple to antiferromagnetism. Indeed, by writing a single ferroelectric domain in the archetype BiFeO3, the number of AF variants is reduced to three, making a quantitative analysis feasible. We have recently achieved to image both ferroelectric and AF domains in a BFO(50nm)/SrRuO3/SrTiO3(001) thin film electrically written using a Piezo-Force microscope. The AF domains reconstructed in a single polarization pad, after a full polarization analysis of the light emitted in every pixel, have typical micron sizes (see Figure: *(left) PFM image of a PFM written ferroelectric monodomain in BiFeO3. (Right) SHG reconstructed AF contrast showing AF domain in the micron range*). This allows to test several external stimuli to attempt writing AF domains, thus paving the way to using insulating antiferromagnets as memory elements.

Importantly, the technique is compatible with time-resolved imaging using pump-probe methods. It consists in sending a pump laser pulse and imaging with a probe pulse arriving with some time delay. The time evolution of the AF domains is thus achievable at a time-scale of the order of the pulse duration, i.e. 100 fs. This should allow us in the near future to probe antiferromagnetic dynamics, which is expected in the THz range.

J.-Y. Chauleau, E. Haltz, C. Carrétéro, S. Fusil and M. Viret, *Multi-stimuli manipulation of antiferromagnetic domains assessed by second harmonic imaging*, Nature Mat. 16, 803 (2017)

Results achieved in the framework of the project ANTIDOX funded by topic emergence and carried out by Michel Viret (SPEC), Stephane Fusil (UmPhy) and Rachid Belkou (SOLEIL).

regime electroluminescence is enhanced by a magnetic field by up to 400% near room temeprature at 7 Tesla fields showing that the control of the spin degree of freedom could have a dramatic impact on SiLED brightness.

Key publication

F. Chiodi, S.L. Bayliss, L. Barast, D. Debarre, H. Bouchiat, R.H. Friend and A.D. Chepelianskii, Room temperature magneto-optic effect in silicon light-emitting diodes, Nature Communication (2017)



(Left) Gas immersion laser doping (GILD) fabrication of SiLEDs. (Right) Near infrared images of active SiLEDs showing an enhancement of electro-luminescence with increasing

ELLIPS (collaboration between NIMBE, LCP, ICCMO and DPC) investigates Radiolysis as a solution for accelerated ageing studies of electrolytes in Lithium-ion batteries. Lithium ion batteries (LIBs) are efficient power sources used for various applications including mobile microelectronics i.e. phones and laptops. However, ageing phenomena causing loss of performance are not yet fully understood. However, these phenomena are a crucial issue related to providing safe and stable batteries.

LIBs are usually composed of an electrolyte, a lithium metal oxide cathode and an anode where the active material is often graphite. Nevertheless, the ageing studies are lengthy, costly, and most often purely qualitative. The study has demonstrated that radiolysis (i.e. the chemical reactivity induced by the interaction between matter and ionizing radiation) is a powerful tool for a short-time identification (within minutes-days) of the by-products occurring from the degradation of a LIB electrolyte after several weeks-months of cycling. Indeed, the highly reactive species created in the irradiated solution are the same as the ones obtained during the charging of a LIB using similar solvents. Radiolysis also enables experiments at the picosecond time scale, giving thus access to reaction mechanisms. Therefore, radiolysis is a promising tool to rapidly simulate the ageing behavior of LIBs and facilitate the development of next generation batteries.

Key publication

D. Ortiz, V. Steinmetz, D. Durand, S. Legand, V. Dauvois, P. Maître, S. Le Caër, Radiolysis as a solution for accelerated ageing studies of electrolytes in Lithium-ion batteries, Nature Communiation 6 (2015)

HOPIR (collaboration between CSNSM, LAC and GEMaC) investigates the role played by defects in opto-electronic properties of perovskites. Defect engineering is applied to hybrid organic-inorganic perovskites (HOP) with 3D and 2D structures, with strong light emitting properties. These materials are promising low-cost alternatives to traditional

semiconductors for photovoltaics and light emitting devices. Point defects are produced in a controlled way by helium ion irradiation in the range 10-30 keV. Strain and defects energy levels will modify the electronic and light emitting properties of the materials. One observes an enhancement of the optical emission at low temperature. This observation indicates that irradiation defects act as active optical centers, essentially in the low-temperature orthorhombic phase as seen in the dependence of the total photoluminescence yield. Another effect of the ion irradiation directly observable is the emission through new excitonic processes. The temperature dependence of the spectra is under analysis and evidences light amplification after ion irradiation at low temperature.



Comparison between primary electron transfers at the electrodes in the electrolytic charge/ageing processes of a Li-ion battery (in LiPF₅ 1 M/diethylcarbonate C₂H₅OCOOC₂H₅ (DEC) solution as a model electrolyte) (left), and after ionization in the bulk during radiolysis with the same medium (right)

2. Biosciences

SPECTRORADIC (collaboration between ISMO and LCP) is devoted to the photo-stability of isolated biomolecules. Complex mechanisms protect biomolecules from light and radiation-induced damages. Photo-reactions mechanisms in biomolecules are the key for understanding their photostability. Photo-fragments of guinine-based alkaloids resulting from the UV-induced dissociation of biomolecules have been studied using tandem mass spectrometry experiments in a quadrupole ion trap. A UV laser induces photo-dissociation and an IR laser (CLIO FEL or optical parametric oscillator) probes their structure by vibrational spectroscopy. These molecules exhibit complex photoinduced processes involving coupled electron and proton transfer reactions. The photo-fragmentation mechanisms are sensitive to the local environment; those observed for the protonated dimer differ from those in the monomer. The results show the formation of a metastable exotic alkaloid monomer, protonated on the aromatic ring, which results from photo-induced proton transfer followed by the dissociation of the dimer. Last, complexation by sulfuric acid has a protective effect on the molecules and results to a photo-stable complex.

Key publications

I. Alata, D. Scuderi, V. Lepère, V. Steinmetz, F. Gobert, L. Thiao-Layel, K. Le Barbu-Debus, A. Zehnacker-Rentien, *Exotic Protonated Species Produced by UV-Induced Photofragmentation of a protonated Dimer, Metastable Protonated Cinchonidine:* Journal of Physical Chemistry A 119, 10007, (2015)

S. Kumar, B. Lucas, J. Fayeton, D. Scuderi, I. Alata, M. Broquier, K. Le Barbu-Debus, V. Lepère, A. Zehnacker: *Photofragmentation Mechanisms in Protonated Chiral Cinchona Alkaloids*, Physical Chemistry Chemical Physics 18, 22668 – 22677 (2016)

SOMATEM (collaboration between LPS, NIMBE and IGR) studies the coexistence of coil and globule domains within a single DNA chain. The semi-flexible highly charged DNA chain may be found either in an extended conformation, the coil, or condensed into a highly dense and ordered structure, the toroid. The transition, also called collapse of the chain, can be triggered in different ways, for example by changing the ionic environment. Using cryo-electron microscopy, individual DNA molecules have been observed one by one, kept separated and confined inside a protein shell, the envelope of a bacterial virus (80 nm in diameter). The collapse transition is not a two states reaction: for subcritical concentrations of polycations (spermine 4+), part of the DNA is condensed and organized in a toroid and the other part of the chain remains uncondensed around. Two states coexist along the same DNA chain. We imaged these 'hairy' globules and described both the global conformation of the

chain and the local ordering of DNA segments inside the toroid (see figure below).



In a cell nucleus, DNA chains are always confined and exposed to low amounts of condensing polycations. These complex intermediate conformations, with coexistence of neighbouring condensed/uncondensed segments, are therefore of the highest interest: by a local tuning of the condensation, they should provide a fine regulation of the activity of the molecule.

Key publication

B. Sung, A. Leforestier, F. Livolant. Coexistence of coil and globule domains within a single confined DNA chain,. Nucleic Acids Res. 44, 1421-7 (2016)

PHYSBIOPS (collaboration between LPTMS, LPS, ISMO and LCF) aims to promote the unification of the physicsbiology interface in Paris-Saclay. Three types of operations have been organized: a biennial international conference, regular topical workshops and a regular seminar series (see list of events below). As of late 2016 these events had brought together as event organizers researchers from 18 different institutes ("laboratoires") in the Paris-Saclay area, and participants from many more.

LUBIOL (collaboration between LIDYL, ISMO, NIMBE and iTEOX) develops a tool for studiing high time-resolved scintillation of biomolecules, induced by ultra-short electron bunches. The DNA damages due to ionizing radiations result

from two different processes: the direct and indirect effects. The indirect effect is well known through pulse radiolysis studies in condensed phase. Direct effects are mostly studied in gas phase and have almost never been investigated by transient technics. An ultra-fast tool has been developped for direct effects studies in condensed phase. The tool is a high repetition rate (76 MHz) femtosecond laser-triggered electron gun, producing bunches of 1 to 200 electrons, with an energy range from 1 to 5 keV and a temporal width from 5 ps to 100 fs. It is coupled to luminescence-decay measurements by Time Correlated Single Photon Counting (TCSPC). The sample consists in solid DNA deposited on a sapphire substrate by a spin-coating technique, covered by a 50 nm gold layer. A light collection system based on a microscope objective working in reflexion is used to guide the emitted photons by scintillation to a PM-MCP C10373 (Hamamatsu) photodetector. Time measurements take between 5 and 30 min. There is no scintillation issued from the golden substrate in absence of DNA.

E-CONNECT BACTERIA (collaboration between LPS, I2BC and ISMO) is devoted to the study of Shewanella oneidensis MR-1. considered as a model of electroactive bacteria and is well-studied for its respiration versatility. S-oneidensis is able to reduce metal ions (like ferric ions) by electron transfer processes or to adhere to electrodes surfaces and to form a biofilm that generates low levels of current. In spite of numerous applications (waste water treatment, biosynthesis of metal nanoparticules, green fuel cell), the exact mechanisms used by the bacteria to transfer electrons to metals remain unclear. The experiments try to link the affinity of S-oneidensis to metals and the current generation. Work is performed on iron, one of the most abundant metal in the soil and on metallic layers that are thin enough to be semitransparent and to allow optical microscopy observations. The layer is placed at the bottom of the samples where the growth medium and bacteria are poured. A rapid increase of the optical transmittance of the sample is observed only few hours after letting the metal in contact with the bacteria medium. The transmittance increases abruptly (figure A) up to unity suggesting rapid and total iron degradation within one hour.



This unexpected result suggests that bacteria convert the solid Iron layer into soluble iron components like in classical corrosion of metals. The rusting of iron involves multisteps of electrochemical process starting by an oxidation of iron thanks to an electron transfer from iron to oxygen. The metal layer has been connected to another solid and "inert" platinum electrode by an external electric wire, the platinum electrode being immersed into the same liquid. The measurement of the electric current potentially generated by the iron oxidation and the flowing in the circuit is performed connecting in series an ammeter. Figure B) shows the current measurement as a function of time. Five to six hours following the liquid inoculation with bacteria, the current rises abruptly up to 20 microamperes before decreasing slowly within the next hour. Both changes in transmittance and electric current are concumitant and clearly related to the microbial biocorrosion.

These two results open new perspectives for basic microbiology applications as well as for biotechnical applications like green bioreactors. Besides the applications, a lot of fundamental work remains to be done to understand the electroactive properties of bacteria, ranging from local measurements and observations (as shown, in Figure C, a preliminary image obtained by phase contrast microscopy) to macroscopic settings with iron foils in combination with a microbiology approach. Genetics tools are under development (construction of mutants Impaired In the phenomenon) in order to better understand and maybe improved the electric current production.

DEEPIM (collaboration between LCF and LOB) is devoted to asset-up for imaging deep inside biological tissues with high resolution. In addition, there is an ongoing trend toward the development of microscopes enabling the monitoring of several parameters on simultaneous independent channels. The imaging depth remains a crucial limitation in heterogeneous and scattering tissues such as the brain and nervous tissues. To overcome this limitation and access to deeper areas, one very promising approach consists in using three-photon instead of two-photon excitation, while shifting the excitation to the SWIR (Short-Wavelength InfraRed) range. Indeed, scattering decreases with increasing wavelength, following a typical $1/\lambda 3$ law. A novel laser source exceptionally suited for this pupose have been designed, providing 70 fs pulses at a repetition rate of 1.25 MHz and with energies in the μJ range. The source was used to image dual-labeled chick spinal cords, and therefore achieve the first demonstration of dual-color 3PE microscopy, along with simultaneous detection of third-harmonic generation signals on a third channel.

SOLARCAPTE (collaboration between ISMO, iBiTec-S, ICMMO and SOLEIL) studies time-resolved interception of multiple charge accumulation in a sensitizer-acceptor dyad. Biomimetic models that contain elements of the photosynthesis are fundamental in the development of synthetic systems that can use sunlight to produce a fuel. The critical task consists in running several rounds of lightinduced charge separation required to accumulate enough redox equivalents at the catalytic sites for the target chemistry to occur. Long-lived first charge separated state and distinct electronic signatures for the sequential charge accumulated species are essential features to track these events on a spectroscopic ground. This work, uses a novel double-excitation nanosecond pump-pump-probe experiment to interrogate two successive rounds of photoinduced electron transfer on a molecular dyad containing a naphthalene diimide (NDI) linked to a [Ru(bpy)3]²⁺ chromophore using a reversible electron donor. An

Highlight 16. Time-resolved interception of multiple charge accumulation in a sensitizeracceptor dyad

Stéphanie Mendes Marinho, Minh-Huong Ha-Thi, Van-Thai Pham, Annamaria Quaranta, Thomas Pino, Christophe Lefumeux, Thierry Chamaillé, Winfried Leibl, Ally Aukauloo

Biomimetic models that contain elements of the photosynthesis are fundamental in the development of synthetic systems that can use sunlight to produce a fuel. The critical task consists in running several rounds of light-induced charge separation required to accumulate enough redox equivalents at the catalytic sites for the target chemistry to occur. Long-lived first charge separated state and distinct electronic signatures for the sequential charge accumulated species are essential features to track these events on a spectroscopic ground. In the present work, we use a novel double-excitation nanosecond pump-pump-probe experiment to interrogate two successive rounds of photo-induced electron transfer on a molecular dyad containing a naphthalene diimide (NDI) linked to a [Ru(bpy)3]2+ chromophore using a reversible electron donor. We report on an unprecedented long-lived two electrons charge accumulated (t = 0.2 ms).



S. Mendes Marinho, H.-H. Ha-Thi, V.-T. Pham, A. Quaranta, T. Pino, C. Lefumeux, T. Chamaillé, W. Leibl, A. Aukauloo, *Time-Resolved Interception of Multiple-Charge Accumulation in a Sensitizer-Acceptor Dyad*, Angewandte Chemie (2017)

Results achieved in the framework of the project SOLARCAPTE funded by topic emergence and carried out by Thomas Pino (ISMO), Winfried Leibl (iBiTec-S), Alli Aukauloo (ICMMO) Pascale Roy (SOLEIL) and Thai Pham (SOLEIL).

unprecedented long-lived two electrons charge accumulated (t = 0.2 ms) have been detected.

See Highlight 16 (project SOLAR-CAPTE by Thomas Pino)

3. Plasma Chemistry

ERACOP (collaboration between LCP, LPGP, and LPP) investigates elementary Reactivity and Analysis for Cold Plasmas. The composition and time evolution of cold plasmas are driven by collisional processes implying neutrals and ions and for some part radical or excited species, for which products and efficiency are poorly characterized. The project develops emerging techniques to determine in a controlled way the elementary reactivity between these species on two instrumental setups at LCP. It also use an innovative high resolution mass spectrometry technique based on the PTR-FTICR method developed at LCP on transportable devices to analyze the compounds generated in cold plasmas, in particular plasmas studied at LPGP for decontamination of gaseous effluents (removal of VOCs and hydrocarbons). The skills of several teams are jointed for a better understanding of these complex media by identifying processes or species of importance for their stability and optimisation.

4. Free Radicals and Molecules Spectroscopy

MACO-GT (ISMO) adresses Large scale open shell Astrophysical molecules. The aim is to contribute to an astrophysics data basis with the CH₂OH radical⁴. The theoretical reconstruction of the high-resolution spectra, in the sub-millimeter range and the experimental device are in progress for the submillimetric spectra. The potential energy surface of CH₂OH·radical presents 8 extrema characterized by the angles \angle H₁COH and \angle H₂COH. There are four equivalent minima with *C*₁ symetry, two local maxima and two absolute maxima absolus with *C*₂/v symmetry at 353 and 1528 cm⁻¹. Computation of vibrational states shows that these states appear in pairs close in energy.

SPACE (collaboration between ISMO and LIDYL) aims at obtaining vibrational and electronic structure data with unprecedented precision on several prototypical molecules of the chlorophyll family in view of apprehending some essential details of the charge separation within the reaction center Photo system II. Photodetachment in cold anions (~10 K) will be applied to the vibrational and vibronic structure of chlorophylls in cold isolated conditions and in the presence of small perturbators such as hydrogen bonding complexes. A bound electron in the cold anion is promoted to a distant orbital of the *dipole bound anion*. Vibrational excitation of the dipole bound anion weakly couples the

⁴S. Saebo, L. Radom and HF. Schaefer III, The weakly exothermic rearrangement of methoxy radical (CH3O⁺) to the hydroxymethyl

loosely bound electron to the detachment continuum (vibrational autodetachment - similar to vibrational autoionisation in Rydberg molecules) causing its detachment. The experimental goal is to obtain excitation spectra by monitoring the electron kinetic energy using the velocity map imaging (VMI) technique.

5. Advanced Instrumentation

CAIMAN (collaboration between DPhIEE and LAC) develops the first steps towards a multi-species atom interferometer for inertial measurements. Cold atom interferometry has allowed in the last decades the development of extremely sensitive and accurate inertial sensors for measuring the gravity acceleration, Earth's gravity gradient or rotations. They appear very promising for a wide range of applications such as inertial navigation, geodesy, natural resource exploration, or fundamental physics. Up to now, cold atom interferometers involve the manipulation of a single atomic species. The project at ONERA, aims to handle three atomic species (⁸⁷Rb, ⁸⁵Rb and ¹³³Cs) simultaneously in the same instrument, would allow to take a big step in the field of cold atomic inertial sensors. A strong point of using different atomic species in the same instrument, compared to a single species experiment, is the ability to increase the number of complementary measurements without disturbance that could arise from spontaneous emission, the lasers manipulating one species having no influence on the other species. The experimental apparatus is in construction. An innovative fibered laser system allowing the manipulation of the three atomic species has been developped and has allowed as a first step the cooling and trapping of cesium atoms. In the short term, the next step will be to cool and trap simultaneously the three atomic species at the same position. This laser setup will be also useful for other experiments at LAC such as ionization of laser cooling of a cesium beam.

FIR HD (collaboration between ISMO and SOLEIL) studies a high density THz frequency comb produced by coherent synchrotron radiation. Frequency combs have enabled significant progress in frequency metrology and highresolution spectroscopy extending the achievable resolution while increasing the signal-to-noise ratio. In its coherent mode, synchrotron radiation (CSR) was accepted to provide an intense terahertz continuum covering a wide spectral range from about 0.1 to 1 THz. Using a dedicated heterodyne receiver, the experiment reveals the purely discrete nature of this emission. A phase relationship between the light pulses leads to a powerful frequency comb spanning over one decade in frequency. The comb has a mode spacing of 846 kHz, a linewidth lower than 200 Hz, and no frequency offset.

radical (CH2OH \cdot), The Journal of Chemical Physics 78 (2), 845-853 (1983)



The figure above shows the CSR extracted by the AILES beamline of SOLEIL and analyzed by 1) a 30 MHz ultimate resolution Fourier Transform interferometer (top panel), and 2) by the new ultra-high resolution heterodyne receiver in a small spectral region around 200 GHz (lower panel). The same heterodyne receiver has been used to generalize this observation to all CSR operation modes at SOLEIL (single bunch, 8 bunches and low alpha). These 3 operation modes emit intense THz frequency combs with different spectral caracterics (in terms of bandwidth, linewidth of each comb, frequency offset) and will be used to perform ultra-high resolution molecular spectroscopy in the THz domain.

Key publication

S. Tammaro, O. Pirali, P. Roy, J.-F. Lampin, G. Ducournau, A. Cuisset, F. Hindle, G. Mouret, *High Density Terahertz Frequency Comb Produced by Coherent Synchrotron Radiation*, Nature Communications, 6, 7733 (2015)

6. Exploratory Projects

CLEPTO (collaboration between LAC and LPTMS) opens the field of the internal temperature of a levitated nanodiamond. Observation of quantum phenomenon on a mesoscopic object requires a perfect control of its environment. An efficient method to gain such a control is to levitate optically a nanoparticle in vacuum. This approach has known important developments, with numerous perspectives, from weak forces metrology to stochastic thermodynamics, through the study of gravity-induced quantum decoherence.



Levitation of a nanodiamond in vacuum: (a) Photography of the experimental setup showing a leviated nanodiamond (white arrow).
(b) Time trace of the particle dynamics. (c) Energy of the centre of mass of the levitated diamond, at a pressure of 2 mbar, as a function of the trapping laser power.

The use of an intense trapping laser for the levitation could lead to an increase of the internal temperature of the levitated nanoparticle. The particle heating will then limit the stability of the system and will impact the centre of mass motion of the particle, as shown in figure (c). This effect could mask quantum effects and will limit force sensitivity of the apparatus. The measure and the control of the internal temperature of the levitated nanoparticle are then crucial for the development of levitation experiments. The levitation of a nanodiamond hosting NV defects, which constitute a sensitive thermometer, is an essential step toward the understanding of the impact of internal temperature on the particle dynamics. It should also provide a solution for a better control of the particle heating.

Topic 5 "Higher Education"

The « higher education » topic focuses on funding actions among master and PhD levels along the reseach LabEx topics. It also contributes to popularization of light and matter physics among the general public, and especially highschool and licence students in France and abroad.

The LabEx plays a specific role among the large higher education network on Plateau de Saclay. Idex Paris-Saclay also contributes to higher education through specific calls (for labs, for innovation in pedagogy, etc). However, Idex mostly funds very large programs with many institutions involved and spanning all science fields, with multi-step heavy procedure among various institutions. One strength of the PALM « Higher Education » is that it consists in a single-step light and simple procedure, involving researcher-teachers within the same research community, and with a pretty good ratio of success (ranging from 50% to 80%). It hence helps funding small-scale projects in an efficient process, very complementary to the Idex fundings and other national large-scale programs. Furthermore, it allows to support original actions, such as forums, schools, trainings, visits which helps make the master degrees more closely connected to the research and industry areas.

The PALM education topic also helps for a better communation and exchange of best practice among the teaching instutions of the Saclay area, first through direct contacts, but also through the discussion of the various proposals, and through the website where each project was documented and openly shared.

On the popularization and attractivity side, PALM also funded a large spectrum of outreach actions described hereafter. These LabEx funding and evenmore the existence of this PALM community appeared as a unique opportunity to develop such popularization items and actions on the specific field of atomic and condensed/complex matter physics. It helps rebalance a little bit the place of atomic and condensed matter physics compared to high energy or astrophysics which already do a great communication work because of large scale institutions (CERN, ESA, NASA…). These actions take benefit from the simultaneous presence of the broad range of expertise among PALM researchers in atomic, condensed or complex matter, together with the expertise of a few researchers in popularization and outreach.

The organization of the topic is based on a "bureau" composed of 9 members, of various research and high education institutions of PALM (Université Paris Sud, Polytechnique, CEA, CNRS, ENS Cachan, IOGS...). The "bureau" is in charge of proposing each year the call for new projects. Once being approved, the various propositions are judged with 2 referees, with possible external referees if needed. The members of the "bureau" are also in charge to advertise the calls in their own institutions and communities in addition to the official LabEx communication.

Since the begining of the LabEx, the topic has funded 19 labs developments, 16 projects about pedagogical innovation and attractivity, 23 schools, and 18 outreach projects, together with PhD and master grants. Its budget consists in 9% of the total LabEx budget. We will focus hereafter only on the 2015-2017 period. In this 2015-2017 period, about half of the budget ($300k \in$) was attributed through yearly calls, the rest being devoted to the organization of LabEx schools, PhD grants, Master grants or a few specific actions.

1. PhD programs

Since 2015, 10 PhD grants have directly been funded by the LabEx, equally shared among all the research axis and decided by the research axis through the call procedure.

See PhD grants

In addition to these grants, various schools targetting PhD students, post-docs and even pre-docs were also organized with the help of the LabEx funding. Some schools followed a traditional framework (talks, discussions, posters), while others involved specific trainings, for example computer simulations at IDRIS center. One was even fully organized and handled by PhD students alone (RJP 2016). In the 2015-2017 period, PALM funded the Beg Rohu Summer School of statistical physics and condensed matter (Quiberon), a DFT school at IDRIS, a school about cristallography, the MOLIM training school (advanced

technics for molecular spectroscopy and dynamics) et Paris-Sud, the conférence ETSF Young researcher in spectroscopy, GDR school on molecular dynamics at IDRIS, and the "Rencontres de Jeunes Physicie·ne·s" RJP.

Most of these schools were organized in the Saclay area. When schools were organized elsewhere, a specific lowcost subscription rate was usually provided to the Saclay students.

The LabEx also devoted a specific budget to organize its « own » schools, one for each research axis. A school will be organized by E. Raspaud (LPS) in sept 2018 about « physical approaches to understanding microbial life » (topic 2), another one in May 2018 about « attosecond dynamics » by P. Salières (CEA) (topic 3), and a third one in 2019 for topic 1 (in preparation). This specific funding is a unique opportunity to gather the LabEx community, especialy its PhD students, but these LabEx schools are open to other researchers as well. The fact that a simple and

PhD Grants

Project: Economique Candidate: Stephan Jennewein Laboratory: LCF Director: Antoine Browaey Date: APR 2014 to MAR 2017 Subject: Study of the cooperative emission of light by an atomic cloud

Project: Deeppaint Candidate: Marguerite Leang Laboratory: FAST Director: Ludovic Pauchard Date: OCT 2014 to SEP 2017 Subject: Penetration of a solvent into a porous gel in consolidation: application to the restoration of works of art

Project: SOC Candidate: Alex Louat Laboratory: LPS Director: Fabrice Bert Date: OCT 2015 to SEPT 2018 Subject: Novel electronic states induced by large spin-orbit coupling

Project: TURBA Candidate: Simon Lepot Laboratory: SPEC Director: Basile Gallet Date: OCT 2015 to SEPT 2018 Subject: Study of the cooperative emission of light by an atomic cloud

Project: FEMTONIC Candidate: Sabuhi Badalov Laboratory: LSI Director: Jelena Sjakste Date: A OCT 2015 to SEPT 2018 Subject: Theoretical study of electronphonon coupling in semiconductor nanostructures and in thermoelectric materials

Project: DRYORDER Candidate: Arnaud Lesaine Laboratory: FAST Director: Georges Gauthier Date: NOV 2015 to OCT 2018 Subject: Amorphous to crystalline transition in films formed by the drying of colloidal suspensions

Project: JOSI Candidate: Chloé Rolland Laboratory: SPEC Director: Fabien Portier Date: JAN 2016 to DEC 2016 Subject: Quantum electrodynamics of a voltage-biased Josephson junction in the high coupling regime

Project: IPAKIS Candidate: Gianluca Aiello Laboratory: LPS Director: Julien Gabelli Date: SEP 2016 to AUG 2019 Subject: Interacting Photons in Array of Kinetic Inductance Superconducting Resonators

Project: SLIDING Candidate: Isabel Gonzales Laboratory: LPS Director: David Le Bolloch Date: SEP 2016 to SEP 2019 Subject: Sliding charge density wave probed by pump-probe ultrafast electron diffraction

Project: IPAKIS Candidate: Mohamed Mafoudhi Laboratory: LSI Director: Nadège Ollier Date: OCT 2016 to SEP 2019 Subject: Rare earth Environnement Modification by Irradiation

sufficient funding is provided by the LabEx makes it much easier and efficient for the local organizers to initiate these schools, as compared to other schools where fundings have to be gathered from many institutions and calls. It also allows to target a specific hot topic close to the research areas of the LabEx.

2. Master programs

At the master level, various pedagogical innovations related to PALM research areas have been supported.

Pedagogical actions were directly supported aiming at developing links between students and research institutions or industry: pedagogical visits at industrial sites for Master PEPs (physique de l'environnement), the student-industry forum for optics at IOGS, visits and seminars for the M2 SBCP (systèmes biologiques et concepts physiques).

Practical labs were funded which propose modern close-toresearch instrumentation and topics : a lab using the LASERIX big facility to measure femtosecond physics, a lab for the measure of Quantum Hall Effect (M2 Nanosciences, cofunded by LabEx NanoSaclay), a lab about surface plasmons (Ecole Polytechnique), one about holography using DMD technology (Ecole Polytechnique), a set of labs about nanomaterials and molecules measuring reactivity together with structural analysis (Paris-Sud), a lab about low temperature extension of LEED measurements (PSud, ENSTA, Polytech, various M2) and a lab about interaction laser-atom through various optical procedures (cold atom physics) (M2LOM, M2 Nano, IOGS), and a high temperature setup for a labs plateform of condensed matter physics.

See highlight 17

The LabEx started a new program since 2015 to fund about 30 grants at master level per year. These grants provide the support to attract interns at the M1-M2 level for 1-3 months stays. A « light » system has been chosen where the grants are distributed among the LabEx labs according to their size and then handled by the labs themselves among their researchers.

3. Popularization of physics

The LabEx has devoted about 20% of its higher education topic to popularization actions, targetting younger students from Licence, prepas, and highschools, but also the general public through various online and live actions.

It supported the French and the International Physics Tournament (FIPT and IPT), a competition about open physics topics for master students from various universities, organized for the first time in France in 2016. Two teams from Saclay area participated to the french tournament (Ecole Polytechnique and Univ. Paris-Sud, who were ranked 2nd and 3rd).

Various medias were developped with the help of the LabEx and then broadcasted through internet (websites, Youtube, Wikipedia...): 3D animations about modern microscopies, a book in french and english about photoemission, a portable exhibit about solid state physics (to come in spring 2018), videos about research and researchers in quantum optics and photonics at IOGS (2018 as well).

See highlight 18

Highlight 17. A condensed matter platform for master labs M. Zeghal, F. Bert, P. Wzietek (Univ. Paris-Sud)

The condensed matter plateform « PMCM » was built by researchers of the LabEx (M. Zeghal, F. Bert, P. Wzietek *et al.*) in order to propose a full set of modern equipment devoted to condensed matter property measurements. It was funded twice by the LabEx, at its start in 2012 and more recently for further high temperature equipment in 2017, together with other Paris-Saclay fundings. It displays a range of state-to-the art equipments to measure both structural and electronic properties with 3 NMR setups, 6 X-Ray diffractometers and fluorescence spectrometers. It allows Master students to learn spectroscopic techniques both on fundamental and applied sides. On fundamental side, it helps better measure and understand diffusion, photoelectric effect, X fluorescence, diffraction methods, nuclear resonance, two-state levels, etc. On material side, it allows to measure various structural and electronic properties of materials. In solid state physics and chemistry, students are for example able to study the ferroelectric transition in perovskites such as BaTiO₃ they have synthesized themselves. In solid state physics, NMR study of minerals is proposed, such as dipolar couplings of water molecules in gypse. In soft matter physics, students can measure the molecular anisotroy of polymers in rubber under mechanical strain.



a few elements of the platform: NMR field, X-ray powder and central diffusion setup and generator, NMR spectrometer, data: a quadrupolar splitting of an NMR spectrum in deuterated rubber under strain

This platform is a nice example of a collaborative setup developed by researcher-teachers from the LabEx and now open to about 400 students from various degrees and institutions: physics magistere or solid state chemistry Master (Univ. Paris-Sud), Polytech, master and magistere at ENS Paris-Sclay, Ecole Centrale-Supelec...

Conferences and live events were organized for the general public and especially highschools and "prepas" during the 2015 International year of light. Small scale animations about statistical physics presented by researchers at science fairs were supported at the LPTMS lab.

See highlight 19

Finally, a large-scale project was initiated by PALM steering committee to explain the physics and research at play in the LabEx: this project made in collaboration with graphic designers, webdesigners, cartoonists and animation videasts consists in a full set of media gathered in a website www.AtomeLumiereMatiere.fr. It proposes a description of the various fields of the LabEx through aesthetic animations about atomic physics, solid state physics, complex matter and nanophysics. It also uses cartoons and drawings to show various research and researchers of the three LabEx axis, focusing on a specific team for each axis with a full interactive cartoon, but also presenting many other research. The aim of this website is first to provide to the general public and to the LabEx present or potential actors a general but entertaining description of the physics at play.

But the project has a broader scope, namely giving a clear description of the atomic and condensed matter physics which can be reused also by non-LabEx researchers, media, etc. Each of the medias presented in the website is therefore available both in french and english and in a downloadable version, and does not explicitely quote the LabEx (except of course in the credits).

In consequence, anyone can use it for other communication or popularization purposes. Possible users are : researchers of the LabEx, but also other researchers of the same physics fields, teachers, popularizers, institutions (Univ Paris-Saclay,

Highlight 19. The international year of light S. de Rossi, N. Westbrook and other researchers from IOGS



conference for highschool students

CNRS, CEA, etc), medias, Youtubers...In addition, various print media were made also available to download for practical use: flyers, postcards, posters... J. Bobroff, responsible for this project, will also use its network and collaborations with science museums and highschool hands-on activities with children

physics teachers associations to spread widely these outreach resources.

Website: www.AtomeLumiereMatiere.fr

Topic 6 "Innovation and Technology Transfer"

The main innovation action concerns a specific annual call organized jointly with LabEx NanoSaclay. The two LabEx also try to promote innovation among researchers by organizing a "Innovation Day" every two years.

That call aims at supporting projects at the upstream edge of innovation and teams that are in first phases of innovation aspects (proof of concept studies, development of pilots...). Indeed, we continuously develop state-of-the-art instrumentation for imagery, metrology, diagnostic, control/command... These innovative set-ups are precious by-products, which can fulfill academic or industrial requirements for other application fields. We deliberately place our actions as close as possible from the LabEx and the starting point of their innovation journey, even assuming some "training" on innovation issues for the teams which are not familiar with ITT (Innovation and technology transfer).

The NanoSaclay/PALM innovation board consists of ten researchers from both LabEx (see annex). It selects and accompanies the most promising projects, providing financial support e.g. for prototype creation or for market studies.

1. Funded projects

A total of 20 ITT projects over 27 have been funded by NanoSaclay/PALM over the last 3 calls (2015-2017), for a total amount of 990 k \in . Eleven of those projects are more relevant to PALM topics and are mainly devoted to instrumentation. A rough classification can be sorted out as follows.

Six projects relate to imaging and filtering devices: plasmonics nano-antennas for terahertz multiband imaging by infrared thermoconversion (Valo-Imhotep), novel system integrating, with the atomic force microscope, a broadband laser with a Michelson device (nanoFTIR), measurement device for the spatio-temporal metrology of ultrashort lasers (EXYT- see highlight 1), imaging of single molecules for biology (NANOBIO), multiscale asynchronous rapid scanning for pump-probe spectroscopy of biological systems (MARS, [SOLINAS 2017]), and one shot THz imaging for non-destructive testing (AMEFI-STRIPP).

Two projects involve the development of sources: a nonlinear achromatic set-up for high harmonic generation (GRADIAN, [GOBERT 2016], tilt interferometry as a tool towards a laser combining coherent laser fibers (PISTIL, [DEPREZ 2016].

One project deals with nanobiotechnologies: development of a system for pathogen nanoparticle-based optical detection (PANOPTIC).

The project DICHRO50mK concerns a technology transfer from a laboratory of the Synchrotron SOLEIL to the Cryoconcept company, to realize a ultra low temperature set-up in ultra-high vacuum environment for XMCD (X ray magnetic circular dichroism) measurements (see highlight 2).

Another project, 3DPAuW, is focused on the elaboration of AuW alloys, initially developed for spintronic applications, to jewelry (see highlight 3).

Regarding PALM ITT funding, most expenses are possible, except salaries of permanent staff: the LabEx provides basic budgets maybe small (50 k€), but very flexible, with a wide eligibility in the expenditures, and with a possibly reoriented nature of expenditure to accommodate the evolution of the action, in a fast way. These qualities were underlined by several project leaders.

2. Innovation Day

The two LabEx organized in October 2015 a meeting between leaders of innovation projects and researchers. The key objective was to promote the different avenues for innovation (licensing, transfer to SMEs, start-up companies...) by examples of successful experience. The first edition of the innovation day began with an introductive talk on intellectual property and interest of filing patents, followed by several contributions from researchers who have participated in the creation of innovative companies (Genewave, Fastlite, Phasics, Biowintech, Nawatechnologies), emphasizing several key points: sources of financing, licenses, organization. These presentations have been complemented by talks given by Paris Saclay SATT (Société d'Accélération de Transfert Technologique)/ SATT). About 75 researchers, engineers, entrepreneurs and students participated to this innovation day. One central outcome of that meeting was the opportunity to share experiences and make valuable contacts.

In June 2017, the Labex PALM and NanoSaclay organized the second edition of this "innovation day". The topic of that meeting was "the researcher as inventor and entrepreneur". It included presentations of six researchers who valued their scientific inventions by creating technological companies and payed tribute to two researchers – entrepreneurs of Paris-Saclay. The meeting, open to young researchers, post-doc and PhD students, gathered around 60 attendees and fostered exchanges between participants.

3. Leverage effect

A possible indicator of the leverage effect would be the number of filed patents linked to PALM projects. However this indicator is difficult to measure, because the maturation time of a technology transfer is larger than the time of the PALM project. Some patents can be filed before the PALM funding, some can be filed a long time after the end of the project, in a context of multiple fundings. And there is no similar easy-to-trace system of acknowledgement as like for publications. Concerning the projects supported through the innovation call, the leverage effect is specific to each situation. Some projects continue mainly by other prematuration or maturation funding (ANR, Idex, SATT). For example, the projects DICHRO50mK and PANOPTIC have been funded by the SATT Paris Saclay, respectively in 2015 and 2016, to continue the maturation process. Some PALM ITT fundings make a technology transfer to an existing company possible, like in the case of the EXYT project, or DICHRO50mK.

Furthermore, it should be noted that, since the last CSI meeting, a new spin-off company has been created in 2016. DAUMET, following NanoSaclay/PALM funding (3DPAuW project). It develops innovative gold materials - new, ecofriendly white gold alloy made out of tungsten - dedicated to the luxury industry (see highlight 2). Another spin-off company, Teratonics (Advanced Terahertz Photonics for Enhanced Productivity and Reliability) has been created in 2016, following two PALM ITT projects : CASIMO-STRIPP (2013) and AMEFI-STRIPP (2015). This company offers solutions for non-destructive testing based on THz spectroscopy. Furthermore, we can also mention that two spin off companies driven by NanoSaclay researchers have just been created: QUANDELA, following the SOPHUC project (efficient single-photon sources for quantum optics), and SPIN ION Technologies, following the homonym project (a novel method for nanofabrication using irradiation by light ions of magnetic materials).

Highlight 20 : EXYT Measurement devices for the spatio-temporal metrology of ultrashort lasers

A. Borot, F.Quéré, J-M. Morchain, O. Gobert (LIDYL), A. Jeandet (LIDYL and Amplitude Technologies)

Ultrashort laser sources have now become major scientific tools, used in very different research fields. The development of these light sources has been intimately coupled with progress in optical metrology. In particular, the ability to accurately measure the temporal evolution of the local electric field E(t) of ultrashort pulses has played a crucial role in the optimization and applications of these lasers. Such measurements, once at the forefront of optical metrology, can be now routinely achieved thanks to a variety of elegant techniques. Yet, all laser beams unavoidably have a transverse spatial extent in the (x,y) plane normal to the propagation direction z, and their complete characterization thus actually requires measurement of the complex-valued electric field E(x,y,t) in space-time.

Fully determining this spatio-temporal electric field is crucial from several respects. First, by providing comprehensive information on the laser beam, it opens the way to unprecedented optimizations of its properties, which is essential for applications requiring the most advanced performances. Second, the interaction of these beams with matter often involves a collective response of the medium to the laser beam, ultimately determined by the full spatio-temporal field E(x,y,t). Measuring this field and using this information as an input for simulation codes would thus open the way to a much finer understanding of such interactions.

The goal of this project is to develop different commercial devices for the spatio-temporal metrology of ultrashort laser beams, building on the expertise we have acquired on this topic in the last 6 years in the context of our academic research. We have now demonstrated two simple and complementary measurements techniques, TERMITES and INSIGHT, which we think can solve this metrology issue for the whole community of ultrashort laser users. They have respectively allowed the first complete spatio-temporal characterizations of a 100 TW laser in 2016 and, in August 2017, of the PetaWatt BELLA laser system, one of the most powerful ultrashort lasers worldwide. A partnership has been established with Amplitude Technologies on TERMITES, and a first commercial product based on this technique, for measurements on collimated beams, will be available by the end of 2018. Thanks to the PALM grant, we have hired a software engineer to program a reliable and user-friendly version of the data processing program of INSIGHT. A prototype will be available in early 2018, which we will use to find an industrial partner for the commercialisation of a device for measurements at focus.



Image: reconstruction of the spatio-temporal electric field of the collimated beam of the PetaWatt BELLA laser (LBNL, USA) from an INSIGHT measurement at focus.

G. Pariente, V. Gallet, A. Borot, O. Gobert and F. Quéré, *Space-time characterization of ultra-intense femtosecond laser beams*, Nature Photonics 10, 547–553 (2016)

 \ll Dispositif et procédé de caractérisation d'un faisceau de lumière $\ensuremath{\scriptscriptstyle \gg}$

F.Quéré, V. Gallet, G. Pariente

Patent application # FR1455472, filed on the 16/06/2014

Results achieved within the project EXYT funded by the ITT topic, and carried out by Fabien Quéré and Antonin Borot (LIDYL)

Highlight 21 : DICHRO50mK Magnetic dichroism in soft x-rays at ultra-low temperature for Single Molecule Magnet study on DEIMOS beamline

P. Ohresser, E. Otero, F. Leduc, J.-P. Kappler (Synchrotron SOLEIL), Ph. Sainctavit (IMPMC), L. Joly, F. Scheurer, B. Muller (IPCMS).

Magnetically bistable molecules, known as Single Molecule Magnets (SMM), are complexes characterized by a slow relaxation of their magnetization at very low temperature. Due to this memory effect of pure molecular origin they have been the subject of intense research efforts as they are foreseen as unit blocks for molecular spintronics based devices, multifunctional devices and for quantum computation applications. Toward this goal SMM are to be organized on surface and to explore their remarkable electronic and magnetic properties in bi-dimensional architectures, only synchrotron radiation based spectroscopies (X-ray Absorption Spectroscopy and X-ray Magnetic Circular Dichroism (XMCD)) can probe such minute amount of material with element sensitivity.

Hitherto their most original quantum properties are to be observed under 1 Kelvin. Hence SOLEIL, through DEIMOS beamline and associates, undertook the construction of a new ultralow temperature insert aimed to reach 50mK (dichro50). With the financial support of the Labex PALM, we have transferred our experience acquired building former cryogenic inserts to CryoConcept*, a company located nearby the Plateau de Saclay and dedicated to innovation in ultra-low temperature refrigerators. The challenge in the conception and the construction of this refrigerator was to comply with the constraints inherent to XMCD experiment: ultra-high vacuum (UHV) environment, enhanced thermal shielding of hosting end-station, high electrical insulation for sample measurement, automation for multi-users, time optimized warming and cooling procedures.

Dichro50 is now in its final phase of Factory Acceptance Test with a first encouraging 60mK measured at the sample holder position. We now anticipate a delivery early 2018 followed by the commissioning phase in order to start the first measurement in March.



* Exclusive license agreement between SOLEIL, the CNRS, the UPMC and the SATT Paris-Saclay

Results achieved within the project DICHRO50mK funded by the ITT topic, and carried out by Philippe Ohresser (Synchrotron SOLEIL).

Highlight 22 : 3DPAuW

From spintronics to luxury, a start-up adventure

C. Deranlot (UmPhy, CNRS/Thales)

From 2007, the spintronics community works to determine the best material to optimize the Spin Hall Effect (SHE). This effect consists to convert a charge current into spin current by spin/orbit coupling without any ferromagnet and any magnetic field. An efficient SHE material has to present a strong spin/orbit coupling and a high resistivity. In 2011, the best candidate was a CuBi alloy (green points on the figure) and AuW alloys seem to be good candidates thanks to their specific configuration: 5d element diluted in 5d matrix (high atomic number Z). Au and W are not miscible at thermodynamic equilibrium but, for the first time, it has been possible to mix them by PVD (Physical Vapor Deposition). The SHE has been measured on these alloys under thin films and the best result has been obtained for AuW13%at. This alloy presents a spin Hall angle of 10%, which means that it converts 10% of an injected charge current into pure spin current.



Figure: Spin Hall Angle for different materials: ratio between spin resistivity and charge resistivity

These AuW alloys have never been obtained before this work so we realized an important work around their characterization and CNRS fileda patent related to the material and its fabrication process[1]. Then, it appears the AuW25%wt. alloy which corresponds to a 18 carats alloy in jewelry presents a specific color. This binary alloy is the whitest gold alloy ever fabricated and it's also shiny. Historically, it exists two processes to obtain a white gold: to mix Au with Ni which entails allergy problems to 20% for the worldwide population, and to mix Au with Pd which is rare and expensive. In both cases, the white gold alloys are mat so the jewelers have to add a thin rhodium layer on jewels to obtain white and shiny jewels. This thin Rh layer disappears with time. The AuW25%wt. alloy, named SpinD Gold in reference to its origin, appeared as a successful substitute to Rh in jewelry. We discussed with a lot of jewelers and watches fabricants to understand how this alloy could be useful for them. We also improved our knowledge around its potential applications and the financial support from PALM and NanoSaclay Labex has been very important for the success of this maturation process.

With a strong value proposition and an important market, the start-up DAUMET, has been created in 2016 to valorize this AuW alloy in jewelry. Today, DAUMET has two activities: a gold coating service on a large panel of matters for luxury (leather goods, jewelry) and the licensing of its process to the high-end costume jewelry actors. For the first time in the world, it's possible to fabricate white gold plated jewels thanks to the DAUMET's process.

[1] CNRS, Cyril Deranlot, patent FR050618, "Alliage monophasique d' or et de tungstène"

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