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HIGH LIGHTS 2019

NÉEL institut

EDITORIAL

Welcome to this year's issue of our Highlights collection !

This 2019 harvest competes with a much broader selection of scientific and technological achievements featured in the 250-page, five-year self-assessment report that we submitted this fall to the HCéRES* Expert Committee who visited the Institut Néel this December. However, the 15 research and outreach endeavours described in this magazine illustrate quite pertinently the diversity and originality of the research activities undertaken at the Institut Néel. They also bear witness to a fruitful complementarity of the classical and the quantum views in condensed matter science, a winning combination reaching back to the early years of the CNRS, our 80-year-old mother institution.

Starting with quite fundamental studies of enigmatic states emerging in strongly correlated matter at low temperatures, be it helium atoms or electrons, you will go on to read about atomic-level studies at the surface or the interface of nano-objects of zero D, 1D and 2D dimensionalities. Thus acquainted with a number of "quantum materials", you will discover their response to coherent optical excitations which reveal their quantum nature, and in particular the possibility of fabricating intricated quantum states, one of the keys to "quantum engineering". You will then be shown how photons may also be involved in creating physical situations escaping the laws of causality (and defying my own understanding!).

The studies of other quantum systems, where mechanical vibrations are paramount, will convince you that relatively large pieces of matter may be brought into their physical quantum ground state by ingenious nano-mechanical schemes involving (again) ultra-low temperatures and microwaves.

Back to the classical world, observing and/or controlling the motion of individual nanometre-size objects designed for specific purposes has become a strategic issue in many fields including cellular biology and medicine, an issue addressed in various ways at the Institut Néel. This may also involve harnessing magnetic field or laser light by micropatterning films or crystals well known for their outstanding magnetic or non-linear bulk optical properties.

The last contribution conveys our ambition to reach out to a more general audience, proposing for example simple experiments that provide ample but disquieting proof that the observations resulting from our physical senses (in this case, vision) may be misleading, but that analysing this bias may teach us a lot about how our brain and eyes work.

Hoping that this set of articles will convince you that the physical world as science "feels" it at different space and time scales may be at least as exciting and beautiful as we feel it through our own senses, I would like to thank and congratulate heartily all the contributors to the present 2019 edition of the Institut Néel's Highlights magazine.

* *High Council for Evaluation of Research and Higher Education (HCéRES)*

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A fluid able to flow through a pipe without friction is a "superfluid". This property is observed at low temperature (below ~2K) in liquid helium. ^{4}He atoms are bosons and superfluidity arises in ^{4}He as a consequence of a Bose-Einstein condensation (BEC) *i.e.* the occupation of the quantum ground state by a very large number of atoms to form a condensate. That is, BEC and superfluidity have a tight relationship. When liquid ^{4}He is confined in extremely tiny channels ("nanopores"), the onset temperature for superfluidity decreases strongly with the diameter of the channels. In order to understand the microscopic origin of this temperature dependence, we have used neutron scattering techniques, and have discovered that a Bose-Einstein Condensation exists in the confined liquid but surprisingly, unlike in the bulk liquid, a BEC exists above the onset temperature of the superfluid.

In bulk liquid ^{4}He, the onset temperatures of both Bose Einstein Condensation (T_{BEC}) and superfluidity (T_λ), are the same: $T_\lambda = T_{BEC} = 2.17$ K. In the pressure-temperature phase diagram for ^{4}He (Fig. 1) the common onset temperature T_λ follows the dashed "bulk λ line". Because ^{4}He is a dense fluid with a BEC, liquid ^{4}He supports well-defined density modes at low temperature, *i.e.* with long lifetimes, in a large range of wave vectors: the phonon-roton dispersion curve predicted by Landau in 1947, and first observed in 1957 using inelastic neutron scattering. The dispersion curve at low wave vector Q corresponds initially to ordinary phonons but it goes through a minimum at $Q \approx 20$ nm^{-1}, and the modes around this minimum are called "rotons". Rotons are the modes which give the most important contribution to the thermodynamics, and the strongest intensity to the neutron scattering signal. The phonon-roton modes cannot decay into lower energy states because single particle and density modes are coupled into a single mode via the condensate. In bulk ^{4}He, the phonon-roton spectrum and its temperature dependence have been extensively measured. The spectrum shows a broadening when the temperature increases. Above T_λ, in the normal phase, excitations are no longer well-defined, leaving only a very broad spectrum. Thus, the appearance of the phonon-roton modes below the temperature T_{PR} is a reliable indicator for the onset of a Bose Einstein Condensation in the fluid.

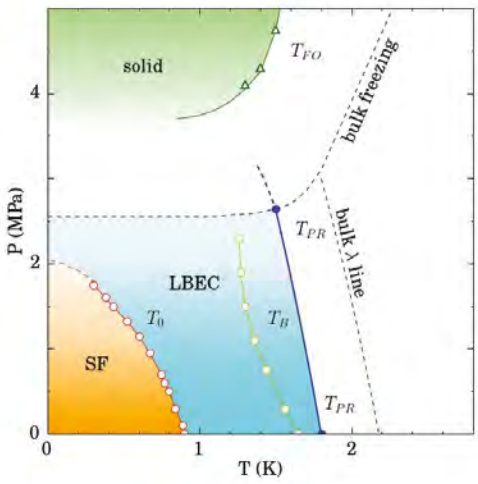
FSM (Folded sheet materials) are a new kind of silica mesoporous materials. They are organized in arrays of ultrafine, straight channels (0.3 microns length), making a honeycomb network, with a narrow distribution of diameters. When liquid ^{4}He is confined in an FSM with 2.8 nm diameter pores, the superfluid onset temperature T_O is lowered from T_λ to the temperature T_O shown in Fig. 1. At pressure $p \approx 0$, the temperature T_O is 0.9 K, and T_O decreases with increasing pressure suggesting superfluidity may vanish at higher pressure in the liquid.

We have carried out inelastic neutron scattering measurements of the temperature dependence of the phonon-roton spectrum of liquid ^{4}He in this 2.8 nm FSM. The experiments were performed at the nuclear reactor of the Institut Laue-Langevin, Grenoble, on the instrument IN5. We have done the measurements at temperatures from 0.45 K to 2 K and at two different pressures. The analysis of the scattering intensity at the low pressure shows that the phonon-roton signal persists above T_O, and becomes extremely broad at 1.8 K; the onset temperature of well-defined modes at $p \approx 0$ is thus $T_{PR} = 1.8$ K. At $p = 2.6$ megapascals there is no superfluidity. However, well-defined modes are observed at low temperature and disappear at $T_{PR} = 1.5$ K, suggesting the BEC exists also at high pressure.

In Fig. 1, we have plotted T_{PR} for our two measurement pressures (0 and 2.6 MPa) and connected them by the straight blue line. Considering that $T_{BEC} = T_{PR}$, our results suggest strongly that we have a temperature range $T_O < T < T_{BEC}$ where there is a BEC but no superfluidity. We interpret this, as localized islands of Bose Einstein Condensation (the region LBEC in Fig. 1) without superflow. The region of localized BEC's is consistent with existing specific heat measurements, the points that are as plotted as "T_B" in Fig. 1.

We propose that superfluid ^{4}He confined in nanopores is an experimental realization of the theoretically predicted "Bose-glass" phase. It is the disorder introduced in the fluid by the walls of the nanopores which is at the origin of localization of Bose Einstein Condensation.

Fig. 1



Pressure-Temperature phase diagram of liquid ^{4}He ($p \leq 35$ MPa) and solid ($p \geq 35$ MPa) confined in the FSM nanopore material. The blue region labelled LBEC is attributed by us to a localized Bose-Einstein Condensate (BEC). This localized BEC region lies between the orange superfluid phase labelled SF ($T \leq T_O$) and the white normal-liquid phase ($T \geq T_{PR}$) in which there is no BEC ($T_{BEC} = T_{PR}$). We observe phonon-roton (PR) modes up to a temperature T_{PR} only, and these modes exist only where there is BEC ($T_{BEC} = T_{PR}$).

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The enigmas of high temperature superconductivity

Superconductivity is a macroscopic quantum state characterised by the absence of electrical resistance below the critical temperature T_c. In 1986, the discovery of superconductivity in "cuprates" (complex copper oxides, most famously yttrium barium copper oxide "YBCO") triggered huge interest. This was not only due to their very high critical temperatures, which rise well above 100 K for certain cuprate materials, but also because they could not be described by the standard Bardeen-Cooper-Schrieffer (BCS) theory of superconductivity which invokes electron pairs ("Cooper pairs") bound together by collective vibrations of the atomic lattice. Unravelling the mystery of the cuprates remains one of the most challenging issues of modern solid-state physics. Just determining the "glue" that binds the cooper pairs in cuprates is a riddle, but the real challenge is to determine the nature of the exotic particles of the normal phase that get coupled into pairs in the superconducting phase. Those particles are not free electrons, not even quasiparticles interacting with the medium, but some kind of "highly entangled mixture" of wave functions.

The complexity (and richness) of the physics of high T_c superconductors is ineluctably related to the complexity of their phase diagrams. In conventional metallic superconductor systems, the superconductivity develops via a "simple" phase of a Fermi-sea of electrons. But high T_c cuprate materials go through a cascade of diverse phases involving both charge and spin ordering as the concentration of holes (the usual electrical-charge carriers) is changed by chemical doping. What makes the situation extremely complex in the cuprates is that the fundamental nature of some of those competing phases remains unknown, especially the very enigmatic, so-called "*pseudogap*" phase.

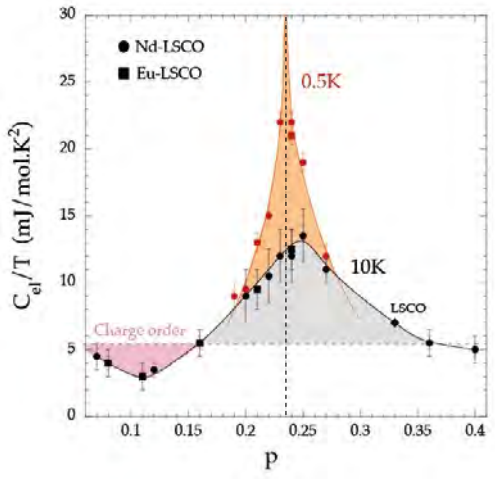
This pseudogap phase seems to play a central role because it vanishes for a hole-doping concentration p equal to a critical value called p^* which yields the maximal critical temperature T_c. It was hence of fundamental importance to determine experimentally the nature of this parameter p^*, in particular regarding whether it marks a "Quantum Phase Transition"(see below). However, no thermodynamic evidence for any kind of phase transition had been observed so far at the onset of the pseudogap phase.

In classical (*i.e.* thermally driven) phase transitions the thermodynamic properties are driven only by the divergence of spatial fluctuations of the order parameter. Whereas, in *Quantum* Phase Transitions (which persist as T approaches zero K), dynamical, *i.e.* temporal fluctuations also play a role as the system is tuned by (varying some parameter) towards the "Quantum Critical Point" (QPC). The transition is then driven by the fluctuations imposed by Heisenberg's uncertainty principle and the divergence of the correlation time leads to a decrease of the energy scales to zero. This gives characteristic signatures in thermodynamic properties, such as a logarithmic divergence of the electronic specific heat (C_e) for T approaching 0.

In collaboration with Louis Taillefer (U. Sherbrooke, Canada), we have measured $C_e(T)$ in $La_{1.8-x}Eu_{0.2}Sr_xCuO_4$ ("Eu-LSCO") and $La_{1.6-x}Nd_{0.4}Sr_xCuO_4$ ("Nd-LSCO") cuprates with doping contents spanning from well below to well above p^* (Fig. 1) We have chosen these relatively low T_c (< 20 K) materials because their superconductivity can be fully suppressed with an accessible magnetic field (~ 18 T). Hence, with magnetic field applied, the doping dependence (p) and the T dependence of the specific heat of the *normal state* could be studied in detail.

As expected in metals, the measured C_e/T was of the order of 5-6 mJ/molK^2 and temperature-independent far from p^* (being equal to the Sommerfeld coefficient). But C_e/T displayed a strong peak for $p \sim p^*$ and T approaching zero, associated with a log (T) dependence (Fig. 1). This is the behaviour known for Quantum Critical Points in *e.g.* magnetic materials.

Fig. 1



Electronic specific heat of two cuprate materials "Eu-LSCO" and "Nd-LSCO", as a function of hole-doping p (holes per crystal lattice unit). As shown, C_{el}/T displays a pronounced peak at a doping content corresponding to the onset of the pseudogap phase for $p < p^* \sim 0.235$ (phase left of vertical dashed line, at right there is a normal metal). The peak is stronger at low temperature (T = 0.5 K, orange area and red datapoints ; T = 10 K, grey area and black points). The doping dependence and the log (1/T) temperature dependence are the classic signatures of the existence of a Quantum Critical Point. Here the parameter driving the phase transition is the doping p. (The dip for p ~ 0.11 (purple area) is formation of a charge-order phase.)

We hence have obtained the first thermodynamic evidence that the pseudogap phase of the cuprate materials ends at a QCP, whose associated fluctuations are then most likely involved in the Cooper pairing and in the anomalous temperature dependence of the resistivity. The nature of those fluctuations is still open, but the existence of a "complex mixture of highly entangled quasiparticles" (instead of standard electrons) in this quantum critical state could be essential to trigger high temperature superconductivity.

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Why do disordered superconductors resist high magnetic fields?

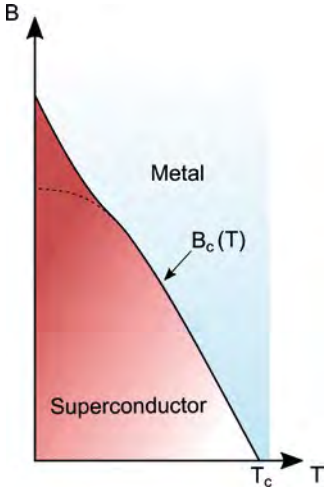
Superconductors are materials that lose their electrical resistance entirely when they are cooled down to low temperatures. In this zero-resistance state, a superconductor exposed to a weak magnetic field expels the field and maintains its superconductivity. This is called the Meissner effect and is explained well by the Bardeen-Cooper-Schrieffer (BCS) theory of 1957. When the magnetic field is increased, however, it eventually begins to penetrate the superconducting material via nanometre-scale, non-superconducting tubes carrying the magnetic flux, the "Abrikosov vortices". At a critical magnetic field B_c, superconductivity is fully suppressed. This occurs when the density of the vortices is such that they fill the entire volume of the material. For usual superconductors, in good agreement with BCS theory, the critical field that suppresses superconductivity increases almost linearly as the material is cooled until it levels off at some low temperature (see Fig. 1) However, for superconductors that are strongly disordered, such as certain alloys or amorphous materials, B_c continues to increase linearly, as the material approaches absolute zero temperature (see Fig. 1). This anomalous behaviour has remained an enigma for decades.

Institut Néel researchers, in collaboration with researchers from the Landau Institute for Theoretical Physics (Russia), the Weizmann Institute of Science (Israel) and the University of Utah (USA), have now lifted the veil on this anomaly. We have studied in particular, as a function of applied magnetic field, the properties of the critical current density defined as the maximum density of current that the superconductor can withstand with zero resistance. Beyond this critical current, superconductivity vanishes and the material recovers its resistivity and energy is dissipated.

We focused our investigations on ultra-thin films of amorphous indium oxide (a-InO), a prototypical disordered superconductor whose amorphous nature produces a strong level of disorder. Our study has involved state-of-the-art instrumentation, enabling us to measure vanishingly small critical currents of a few nano amperes on ultrathin films (30-60 nm thickness) of tenths of microns width, under extreme conditions (down to -273.05°C, *i.e.* T = 0.01 K, and up to 14 teslas magnetic field). These measurements, new for this field of research, have required a stringent filtering and isolation against external electromagnetic perturbations.

An unexpected behaviour of the critical current density was revealed: the critical current density varies in a very simple fashion with the difference between the critical field B_c and the applied field B, according to a power law of exponent 3/2 with high accuracy. This exponent 3/2 was the key to understanding the continuing linear increase of B_c upon decreasing the temperature towards absolute zero. It indicates that the superfluid density ρ_s, (the density of electrons condensed into the superconducting state) varies also as a power law of that difference with an exponent accurately equal to 1 (*i.e.* linear variation $\rho_s \propto |B\text{-}B_c|$).

Fig. 1



Magnetic field vs absolute temperature (B-T) phase diagram for superconducting materials. As T decreases below a superconductor's critical temperature T_c the critical field for destroying superconductivity, $B_c(T)$, increases linearly from zero. The BCS theory predicts a levelling off of $B_c(T)$ at some low temperature (see the dashed curve). But strongly disordered superconductors resist the field, anomalously. The field Bc needed to suppress the superconductivity of these materials increases linearly (the solid black line) down to $T = 0$.

Our team has shown theoretically that this behaviour, together with the specific thermal fluctuations of the Abrikosov vortices in the disordered superconductor, explains the anomalous linear increase of B_c upon approaching zero temperature. Due to the strong disorder in these films, the Abrikosov vortices form a disordered lattice called a "vortex glass". Theory predicts that the thermal fluctuations of these vortices around their pinning centres lead to a suppression of the superfluid density ρ_s (*i.e.* the superconductivity) at a critical temperature $T_c(B)$ that is proportional to the zero-temperature superfluid density. Because our critical current measurements showed that this density scales with $|B\text{-}B_c|$, we can conclude that $T_c(B) \propto \rho_s \propto |B\text{-}B_c|$, which describes the anomalous linear increase of $B_c(T)$.

This work provides the first theoretical model, experimentally confirmed, which describes the anomalous, continuing linear increase of the critical field down to absolute zero temperature as seen for this type of superconductor.
Disordered superconductors are promising materials for quantum information circuits, in particular for hybrid qubits combining disordered superconductors and aluminium technology. The disorder indeed endows the superconductor with a "super-inductance" which is a key element in new qubit designs immune to external electromagnetic perturbations. The protection of the fragile qubit intrication at the basis of the quantum information is an indispensable need for setting up superconducting quantum circuits for quantum computers.

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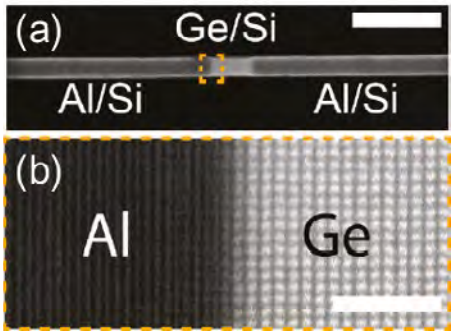
Atomically precise interfaces reveal exceptional quantum effects

Hybrid superconducting-semiconducting systems are promising candidates for nano-electronic quantum devices including quantum bits ("qubits") and quantum circuits. However, their successful realisation requires devices with very few defects. In these hybrid devices, significant defects result from poor quality interfaces between the superconductor and semiconductor. Reducing defects by improving the interface provides a significant challenge.

Fig. 1



(a) Scanning Electron Microscope image of the core/shell (Al-Ge/Si-Al) nanowire heterostructure after annealing. The Al contact pads are out of picture. Scale bar is 100 nm.

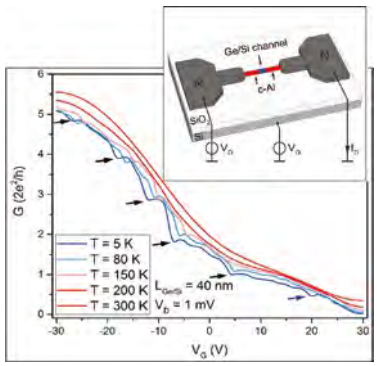
(b) "HAADF" Scanning Transmission Electronic Microscopy image of the Al-Ge interface of the yellow dashed square of (a). Al and Ge atoms are visible with zero defects at the interface. Scale bar is 2 nm.

In collaboration with the Institute of Solid State Electronics at TU Wien, Vienna, Austria, and the Laboratoire d'Etude des Matériaux par Microscopie Avancée, CEA-Grenoble, we have fabricated and characterised hybrid nanowire heterostructures having extremely high quality interfaces. Developed in Vienna, a novel technique facilitates the fabrication of superconducting-semiconducting hybrid nanowires (NWs) with atomically precise interfaces. We start with a 3 micron long nanowire consisting of a germanium (Ge) core with a diameter of 30 nm and a silicon (Si) shell with a thickness of 3 nm. Then we apply two aluminium (Al) contact pads onto the two ends of the wire, using electron beam lithography. Through annealing, we can enable and control the replacement of germanium of the nanowire by aluminium from Al pads. The controlled substitution of Ge by Al results in a monolithic nanowire having the form of a short, central Ge segment, of desired length, connected on both sides by monocrystalline aluminium (c-Al) nanowire segments serving as leads, while maintaining its Si shell (Fig. 1a and inset of Fig. 2). A substantial breakthrough of this method is the atomically precise contacts between the Al and the Ge, which we can observe using a tunnelling electron microscope (Fig. 1b).

To highlight the high quality of these devices and their quantum phenomena we carried out temperature dependent electrical measurements on a nanowire with a Ge segment of length 40 nm between the two Al contact leads. By applying a voltage difference (V_D) across the device and measuring the current (I_D), we can calculate its conductance (the inverse of its resistance) here $G = I_D/V_D$. As germanium is a semiconductor, we can control its conductivity, its ability to conduct charge, by applying an electric field, the gate field. The electric field is induced by applying a voltage bias (the gate voltage V_G) to the highly doped underlying silicon wafer, see the inset of Fig. 2. Germanium in its natural state behaves like a *p*-type semiconductor, so it is easier for positive charges (holes) to transverse the Ge segment. As such, as shown in Fig. 2, applying a more negative gate voltage increases the conductivity of the device.

As we decrease the temperature of the sample from 300 K, we begin to observe structures within the conductance curves at temperatures as high as 150 K. These structures become more pronounced as the temperature is reduced to 5 K. The structures, indicated by black arrows (Fig. 2), are known as quantized conductance plateaux and are a phenomenon of "one dimensional" nanostructures explained by quantum mechanics. The plateaux conductance is then given by two fundamental constants, *e* the electron charge and *h* the Planck constant: $G_n = n2e^2/h$ where *n* is an integer. The ability to observe such features up to 150 K demonstrates the high quality of the sample and its interfaces. From the measured conductance of the plateaux, we estimate the transparency of the Al to Ge interface to be greater than 96%.

Fig. 2



Conductance (G) vs gate voltage(V_G) for fixed device voltage V_D = 1 mV, for the nanowire heterostructure having a 40 nm length Ge segment, measured at different temperatures. Black arrows indicate the conductance plateaux. The inset schematic is the sample under measurement.

Measurements below 1.2 K reveal that the aluminium contacts become superconducting, resulting in the connected Ge segment experiencing *proximity-induced*, superconducting transport properties, such as dissipationless current.

We have demonstrated the exceptional electrical transport characteristics of these high quality core/shell (Al-Ge/Si-Al) nanowires fabricated using a novel annealing technique. Having overcome the limitations of interface defects, these devices can be integrated into quantum devices and used for future investigations of superconducting-semiconducting phenomena.

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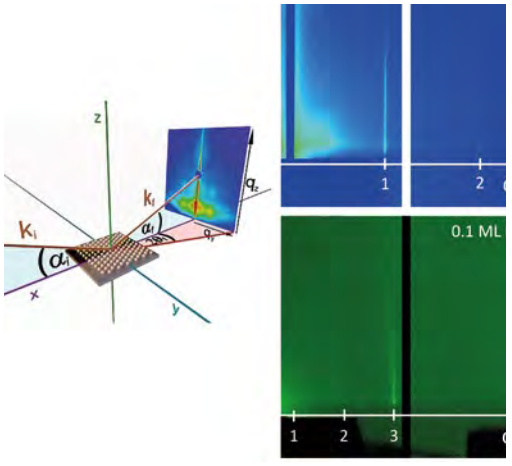
Towards single-atom catalysis: less is more

Clusters containing a small number of atoms can exhibit unique chemical properties. They are very interesting for catalysis as the addition or removal of even a single atom can induce a change in the cluster's chemical activity. Single-atom catalysis constitutes a new exciting frontier in the field of heterogeneous catalysis and is now becoming accessible thanks to the development of new catalyst preparation methods and powerful characterization techniques. One of the most used catalysts in industrial processes and in diesel-vehicle catalytic convertors is platinum (Pt), which is among the most expensive catalyst materials. So the interest of single atom catalysis becomes evident both from an economics point-of-view and regarding the preservation of natural resources. The possibility to grow ordered arrays of tiny monodisperse Pt clusters can pave the way to a better understanding of the link between atomic-scale properties and catalytic performance.

Typical catalysts consist of nanoparticles (NPs) of noble or transition metals dispersed on an oxide powder. As early as 1987, Japanese researchers evidenced a significant catalytic activity for gold nanoparticles, even though gold is totally inert in its bulk state. This triggered a wealth of fundamental studies aimed at understanding how catalytic performances depend on the nanoparticle size. Approaches involving size-selected clusters with a similar shape and a narrow size distribution are of great importance for such studies. Ordering such an assembly of homogeneous nanoparticles in two dimensions would provide even better model systems for studying the correlation between the active site geometry and the catalytic properties (compared to randomly distributed nanoparticles). At the Institut Néel, we are working towards an ultimate model system which would consist of a collection of supported, isolated, single active atoms organized in two dimensions.

Nanostructured substrates are widely used for growing homogeneous assemblies of ordered nanoparticles. Several studies have focused on a specific substrate consisting of an aluminium-oxygen bilayer of atoms obtained by oxidation of the surface of a $AlNi_3$(111) single-crystal. This ultra-thin oxide layer exhibits two hexagonal superstructures having 2.4 and 4.1 nm-periodicity, called respectively "network" and "dot" structures (Fig. 2). These structures can act as nucleation centres for the growth of periodic arrays of metallic nanoparticles.

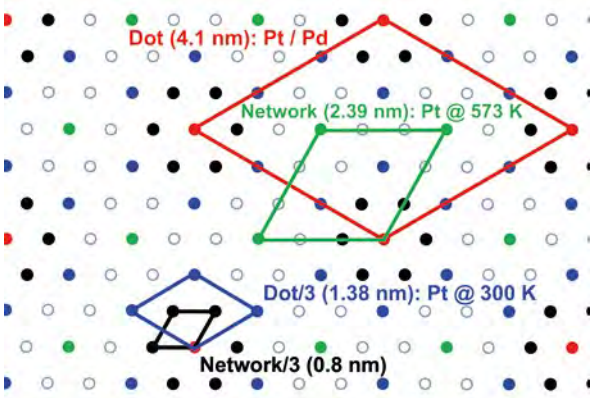
Fig. 1



Grazing Incidence Small-Angle X-ray Scattering pattern of the Al_2O_3 /$AlNi_3$(111) surface seeded by a 0.12 monolayer of palladium.

The degree of order depends critically on the growth kinetics, calling for characterization during nanoparticle synthesis. Grazing Incidence Small-Angle X-ray Scattering (GISAXS) is a powerful tool for such investigations (Fig. 1). In a collaboration with the Interdisciplinary Centre of Nanoscience of Marseille (CINaM) we investigated whether ordered arrays of platinum nanoparticles could be obtained on this specific aluminium oxide substrate. We determined the conditions for Pt growth using GISAXS *in situ* at the French beamline (CRG-IF) at the European Synchrotron Radiation Facility (Grenoble).

Fig. 2



The different superstructures existing at the surface of Al_2O_3/$AlNi_3$(111). The main structures are called "dot" and "network" structures shown at top (periodicity of 4.1 nm and 2.4 nm). When Pt atoms are deposited on this surface, they self-organize on one of these superstructures, but also on a new, smaller structure dot/3 (blue) of 1.38 nm-periodicity. The preferred superstructure depends on the Pt growth conditions indicated in each case.

At 300 K and for a very low coverage (equivalent to ~0.1 monolayer), the platinum atoms organize on a new surface superstructure with a periodicity three times smaller (1.38 nm) than the periodicity of the well-known "dot" structure, leading to a dense collection (~ 6.10^{13} cm^{-2}) of very small clusters of size 1 to 6 atoms (Fig. 2). On increasing the substrate temperature to 573 K, the Pt clusters organize on the "network" structure. If the surface is seeded by a small amount of palladium (Pd) atoms before platinum deposition, larger, bimetallic clusters of ~100 atoms size are obtained. They are organized on the "dot" structure and remain stable up to 733 K. Three types of sites with specific adsorption energies for Pt and Pd thus lead to different organisations (Fig. 2).

This study demonstrates for the first time the possibility of growing tiny clusters of a very small number of platinum atoms in a dense and organized manner on an oxide surface template. This system can be used as a model catalyst for testing relevant catalytic reactions. One key point to elucidate is the stability of the nanoclusters when exposed to real conditions, such as ambient gas pressure or high temperature.

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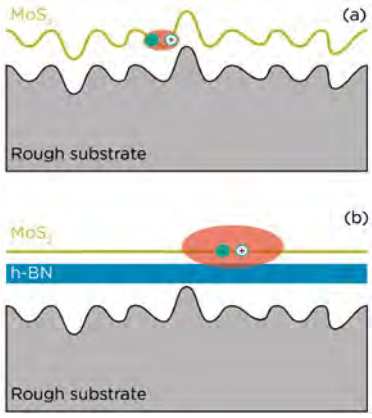
Two dimensional materials get flat and clean

As opposed to bulk materials, two dimensional materials are by construction very sensitive to their environment. This is both a "blessing and a curse". They can be studied directly using ultra-sensitive surface-science techniques such as Scanning Tunnelling Microscopy to reveal effects that are otherwise inaccessible for buried materials. But at the same time, being exposed to their environment can severely degrade their electronic and optical properties. Recent research in the field has led to a better understanding of how to protect these materials, in order to observe their true, intrinsic, properties.

After the (re)discovery of graphene in 2004, which led to the Nobel prize of K. Novoselov and A. Geim in 2010, researchers have applied the same recipe, called "exfoliation", to obtain ultrathin flakes of a wide variety of layered materials, especially to isolate single layers of molecules. Among this very large family of materials, some members, such as molybdenum disulphide (MoS_2), stand out for their unique optical properties. They are thus very promising for designing new optoelectronic devices with enhanced functionalities such as flexibility and greatly reduced size.

Being only a few atoms thick provides opportunity but at the same time any imperfection in its environment will affect the single layer. When the environment is not perfectly controlled it will necessarily vary from one location to another. The single layer's electrons will feel a spatially varying disorder potential limiting the optoelectronic properties of the material.

Fig. 1



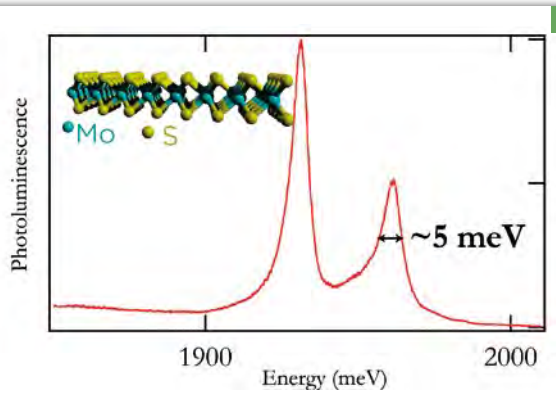
Localised and delocalised excitons in MoS_2.
(a) For a monolayer of MoS_2 lying on a rough substrate, its «excitons» (electron-hole pairs) will be localized in a disordered electronic potential.
(b) With a layer of hexagonal boron nitride inserted, the MoS_2 monolayer does not feel the roughness of the substrate. The exciton can occupy a large space and emit a photon more efficiently.

In order to characterise this effect and to overcome this issue, we have used a spatially resolved, nonlinear optical technique, called four-wave mixing micro-spectroscopy, that can disentangle the intrinsic and extrinsic optical properties. We have observed that if no special care is taken (for instance using a standard silicon dioxide substrate to support the monolayer of MoS_2), the layer's optical properties are dominated by disorder. More precisely, elementary excitations of the systems that are relevant for optical properties, i.e. electron-hole pairs ("excitons"), are spatially localized in this disordered potential. They cannot "spread out" as much as they could wish to (Fig. 1(a)). Because of this localization, the light-matter interaction is reduced, lowering the material's luminescence efficiency, i.e., the rate at which excitons are converted to external photons.

On the contrary, we have found that if the monolayer is protected by putting a thin (about 10 nm thick) layer of hexagonal boron nitride (hBN) over the substrate, the spatial extension of the exciton is no longer limited by the disorder (Fig. 1(b)). This results in clean, intrinsic properties that are otherwise concealed.

A stack of three layers hBN-MoS_2-hBN is called a van der Waals heterostructure as they are held together only by weak, van de Waals forces. In the optical spectra of such heterostructures, we observe strong, narrow emission peaks in the red spectrum having width of order of only a few meV (Fig. 2). This is the signature of properties dominated by intrinsic-material characteristics, no longer masked by the inhomogeneity of the substrate potential.

Fig. 2



Narrow width of the emission peak in the optical spectrum of a high quality MoS_2 monolayer. A monolayer of MoS_2 (molecular model top left) is a direct bandgap material. Electrons and holes are strongly bound in the form of excitons that can emit light efficiently. Linewidths of order 5 milli-electronVolts are observed in a photoluminescence experiment and are the signature of high quality material.

Using our four wave mixing microscopy, we identify areas of order several microns where the optical response is totally dominated by the "homogeneous" broadening, determined only by the excited state lifetime. When not affected by disorder, an exceptional photosensitivity is achieved. This is one of the properties that makes single layers of transition metal sulphides and selenides so promising for future optoelectronic devices. Future work should prove that such van der Waals heterostructures, retaining the intrinsic properties of the active material, can be produced on a wafer scale, making a viable approach for industry applications.

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A quantum optical filter based on semiconductor exciton-polaritons

Realising advanced quantum states of light with visible-spectrum photons in a simple and practical way has become a key goal in the context of expanding quantum technologies. Semiconductor-based, nanometre scale structures are very promising in this context. For instance, semiconductor "quantum dots" embedded in optical microcavities, have reached an impressive level of development, with the demonstration of increasingly complex quantum states of light, prepared with an increasing fidelity. But due to different technological challenges, a strategy that would not require semiconductor quantum dots would be highly beneficial. The mechanism of "quantum blockade" offers such an opportunity.

Originally investigated in the context of nonlinear optics in atomic gases (Rydberg atoms held in an optical trap), quantum blockade consists in engineering strong two-body interactions between electronic excitations, such that the light propagating in the medium experiences an optical nonlinearity so large that it is active down to two photons. In a semiconductor material the best way to do this is to use "exciton-polaritons" in which an electronic excitation (an "exciton", *i.e.* a photo-excited electron bound to a valence-band hole) is in the strong coupling regime with light in an optical microcavity. Exciton-polaritons exhibit a strong two-body interaction via Coulomb interaction, involving their excitonic component.

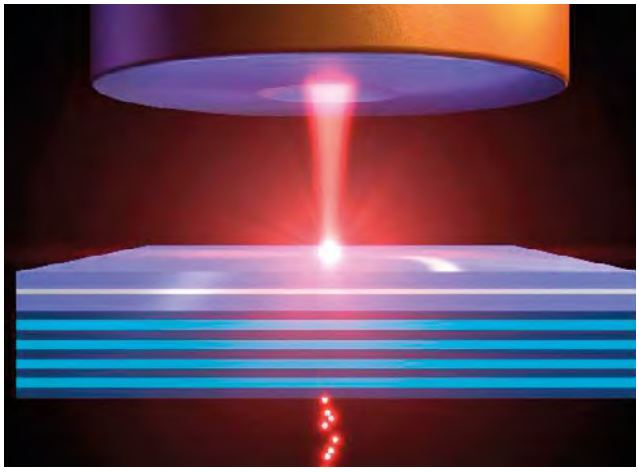
The resulting polaritonic quantum blockade provides an effective "quantum filter" where, when a laser beam is tuned at resonance with the one-polariton transition, the first photon will create a polariton, but the next one will be blocked from entering the system because the polariton already-present has shifted the transition frequency. In practical terms, this regime is achieved when g, the magnitude of the nonlinearity, is comparable with the polariton loss rate γ.

The main challenge researchers faced, in trying to implement this approach, was that even with the most favourable semiconductor material, namely Arsenide alloys, g and γ are hard to match. However, Institut Néel researchers working in a collaboration with the Centre for Nanoscience and Nanotechnology (Saclay) and with the group of Thomas Volz (Sydney, Australia) reported the first signatures of polariton blockade, simultaneously with another team, that of Ataç Imamoglu (Zurich), in February 2019.

To increase the interaction strength, both teams adopted a similar strategy that consists in confining polaritons in the smallest possible volume without compromising on the polariton loss rate γ. To do so, the confinement was realized optically, using a confocal optical cavity, in an optical fibre, featuring a mirror radius of curvature of a few tens of microns (Fig. 1). Experimentally, the polariton blockade regime is probed by driving the system resonantly with a laser, and by measuring the time correlations between two photons in the transmitted beam. With the right tuning of the experiment's parameters, one expects "anti-bunching" in the transmitted photons, *i.e.* a signature of the increased likelihood of the one photon state. Both teams have reported anti-bunching up to 5%, which is sufficient to demonstrate the onset of the polariton-blockade regime.

The present results emerge from a long quest and they make a decisive advance in the nascent field of "quantum polaritonics". They demonstrate that the vast possibilities offered by interacting single-polariton states for processing quantum optical information "on-chip" are realistic prospects. Additionally, this work provides an upper limit on the quantitative estimate of the exciton–exciton interaction strength in Arsenide-based nanostructures, which has been the matter of a long-lasting debate. Now, with a quantitative knowledge of where we are in the quantum regime, realistic strategies are envisaged to increase further the nonlinearity, such as by tighter optical confinement, to get deeper into this quantum regime.

Fig. 1



Artist's image of the experimental setup: The incoming, classical, laser light beam travels through the optical fibre (top of the image) and enters the cavity (red beam). The concave indent (at the centre of the flat surface of the fibre's end) is the cavity's top mirror, a spherical mirror of a few tens of microns of curvature. The flat, bottom structure consists of a semiconductor quantum well (white layer) on top of a Bragg mirror, constituted of alternating semiconductor layers (dark blue/cyan). As a result of the polariton blockade effect, the transmitted light is emitted as separated photons (bottom of the image).

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Realising quantum superpositions of cause-and-effect relations

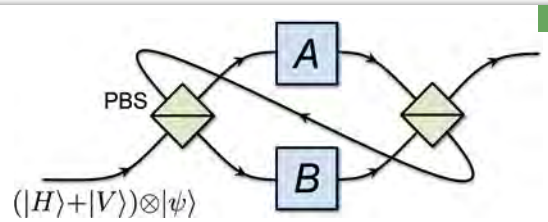
In our everyday life, we are used to understanding causal relations between events in terms of causes and effects, with causes necessarily preceding effects. It has however been realised recently that some new types of causal relations could be found in the quantum world, in situations where no well-defined causal structure can be defined. Indeed, causal relations themselves can be subject to quantum indefiniteness, and put into quantum superpositions.

A remarkable feature in the quantum world is that physical systems sometimes fail to have well-defined properties: a particle can be "both here and there at the same time", and Schrödinger's ill-starred cat can be "both alive and dead at the same time". More rigorously speaking, we say that objects can be in a *superposition* of different incompatible states.

One may wonder if this central notion of superposition is restricted to the states of quantum objects, or could also be applied, more exotically, to cause-and-effect relations. In the classical world, given two elementary events *A* and *B*, one has either that *A* causes *B*, or vice-versa (or that they are not directly causally related). Could we imagine some situation in the quantum world, where (in some sense) "both *A* causes *B* and *A* causes *B*, at the same time"—i.e., in a superposition?

The answer is indeed positive. A paradigmatic example, known as the "*quantum switch*", is a small quantum circuit (see Fig. 1) implementing two quantum operations *A* and *B*, whose order is controlled by the state of a qubit (a 2-dimensional quantum system): if that state is |0>, then *A* is applied before *B*; if that state is |1>, then B is applied before *A*. By preparing the control qubit in a superposition |0>+|1>, one ends up with a superposition of the two different causal orders between *A* and *B*.

Fig. 1



A photonic implementation of the "quantum switch": the polarisation (horizontal, |H> or vertical, |V>) of a photon controls its trajectory through the above setup (via polarising beam-splitters (PBS) which transmit |H> and reflect |V>). An |H> photon first goes to A and then to B, and vice-versa for a |V> photon, where A and B are quantum operations applied to the state |ψ> of another degree of freedom of the photon (e.g., the orbital angular momentum, as in the Brisbane experiment). A photon in a superposition |H>+|V> thus undergoes a superposition of the two orders AB and BA.

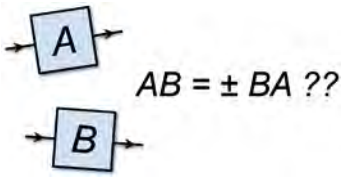
The property that the operations *A* and *B* in the quantum switch do not abide by a well-defined causal structure has been formalised properly, and is known as "*causal nonseparability*". In collaboration with partners in Vienna (Austria) and in Brisbane (Australia) we have shown theoretically how one could demonstrate this causal nonseparability experimentally, by measuring a so-called "*causal witness*". In practice, this amounts to implementing different operations *A* and *B* in a quantum switch, measuring the state of the output system, and combining the statistics so as to estimate a certain quantity *S*, constructed in such a way that well-defined causal relations can only give $S \geq 0$. If experimentally one gets $S < 0$, then for sure there cannot be any well-defined causal structure.

We contributed to an experiment performed in the research group of Andrew White in Brisbane, which realised such a quantum switch in a photonic setup (precisely as in Fig. 1), and measured the value of a causal witness to be negative and 18 standard deviations below the definite-order bound of $S = 0$, thus clearly demonstrating, as explained above, that the operations had no well-defined causal order.

Quantum processes with indefinite causal orders, as in this first proof-of-principle experiment, could in the future have practical applications. Indeed they could realise certain tasks, which are impossible to realise with standard, "causally-ordered" quantum circuits: an example is given in Fig. 2. Or they can perform better than standard quantum computers at certain quantum computation or communication tasks. While the quantum switch is the first known example of such processes, we are currently investigating new types of quantum circuits with indefinite causal order, and the new avenues for possible applications that they open.

More fundamentally speaking, such studies raise interesting metaphysical or philosophical questions about the notion of causality in the quantum world, about the relations between causal structures, space-time structures, and the other central notion of quantum entanglement.

Fig. 2



A possible application for circuits with indefinite causal orders: one is given two back-box devices realising some quantum operations A and B with the promise that these either commute (AB=BA) or anticommute (AB=-BA), and one has to determine which of the two options is true. It is impossible to perfectly discriminate the two cases if one applies A and B in a well-defined order; while this becomes possible by plugging them in the quantum switch (Fig. 1), and applying them in a superposition of the two orders.

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Nano-mechanics near zero temperature

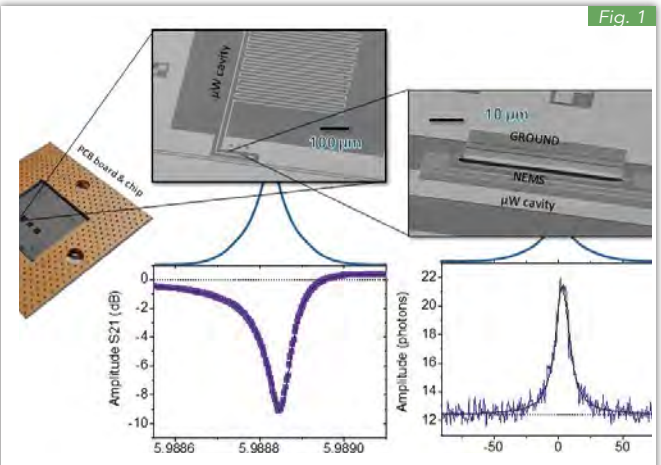
Profiting from clean-room facilities, it is possible to fabricate mechanical elements with dimensions smaller than a micron, in particular vibrating "nano-electro-mechanical systems" (NEMS). As these objects have a set of electrical contacts, they can be embedded in electronic circuits and now even into *quantum* electronics circuits. Using microwave technologies together with a NEMS, the concepts of opto-mechanics can be applied at much lower frequencies (gigahertz), leading to a new type of system perfectly compatible with both state of the art cryogenics and quantum processing. But also, beyond applications as a new resource for quantum electronics, profound quantum concepts can be addressed when NEMS mechanical modes are cooled down into their quantum ground state at temperatures close to absolute zero.

Theoretical proposals to associate microwaves and nano-mechanics for quantum information processing are flourishing. These proposals discuss, among the most popular issues, peculiar microwave circuits and amplifiers, quantum information storage, and coherent photon transfer from microwaves to optics and vice-versa. Some fascinating experimental realisations have already been achieved, but in most of them the quantum limit of these new components is achieved by laser-beam cooling: the mechanical mode is actively cooled (but not its environment) to the ground state by "optical pumping" techniques. Laser cooling has been used because the most advanced technology concentrates on devices having mechanical resonance frequencies around 10 to 20 MHz, so they still host about 10 thermal phonons at the base temperature of commercial dilution cryostats, about 10 milliKelvin.

Other theoretical proposals have explored the limits of quantum mechanics, and its boundary with classical physics. These proposals deal with entanglement and decoherence of non-classical mechanical states. It has even been proposed that NEMS could detect quantum gravity effects. But for testing most of these proposals, the control of the environment that is coupled to the mechanics is a key: out-of-equilibrium situations are prohibited, and the whole system (i.e. not just the mechanical mode but also its environment) should be ideally cold. Therefore active cooling schemes are not sufficient, and a cryogenics facility reaching the lowest possible temperatures is required.

Ground-state cooled nanomechanics is at the core of the European Research Council's Consolidator Grant "ULT-NEMS" (Ultracold Nanomechanics). As a principal participant, the Institut Néel has established a unique platform combining two state-of-the-art technologies: microwave electronics and microkelvin temperatures. The key element for first demonstration purposes is to monitor the temperature of both the environment and the mechanical mode *on-chip*. We have demonstrated the capabilities and the methods in a recent article (see further reading) by means of a conventional microwave setup: an on-chip microwave cavity coupled to a beam-shaped NEMS (Fig.1). Beyond the technical achievement, we also report the intrinsic properties of the device; it happens that a technological limitation for such devices does not lie in thermalisation as is commonly believed, but in an internal instability never reported before, which is still not understood. We demonstrated recently that the same mechanism exists, to a lesser extent, in a "drumhead" (membrane) NEMS. Fundamental quantum studies will certainly be required for understanding this unexpected feature, and to find a way to circumvent it in the future.

Fig. 1



Scanning Electron Microscope images of the microwave cavity (at top left), and of the beam-shaped nano-electro-mechanical system (NEMS) at right. A cavity resonance peak (around 6 GHz) and a NEMS resonance (around 4 MHz) are also displayed.

Our cryogenics and setup is now part of the Infrastructure "EMP" (European Microkelvin Platform), a consortium of 17 European partners sharing expertise and equipment in ultralow temperature physics, giving access to this new frontier of condensed matter physics. The Institut Néel has made its large set of laboratory-built mK refrigerators and its 100 micro-Kelvin nuclear-demagnetisation cryostat available to the EMP partners.

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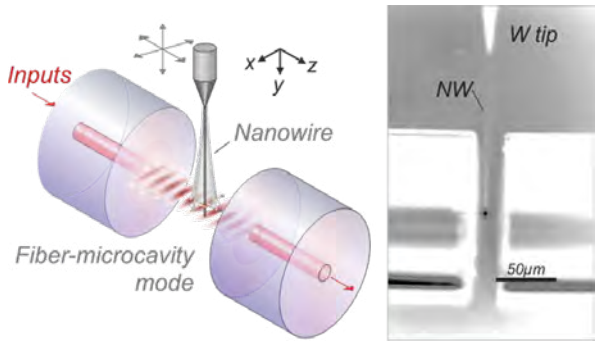
Cavity nano-optomechanics in the ultra-strong coupling regime

Coupling a mechanical oscillator to a probing light field is a key implementation to explore the quantum limits of ultrasensitive, continuous measurements and the inherent quantum "back-action" associated with the measurement process. That is, the light field that serves to readout the mechanical vibrations also exerts a fluctuating optical force, which perturbs the dynamics of the oscillator under investigation. An experiment involving a highly sensitive force sensor, a suspended nanowire inserted in the confined mode of a high finesse optical micro-cavity, shows that one can enhance the opto-mechanical interaction up to the point where the action of a single photon in the cavity mode should become mechanically-detectable.

In collaboration with the Kastler Brossel Laboratory, Paris, we have explored the interaction (in vacuum) between the vibrations of a Silicon Carbide nanowire (100 µm long, 130 nm in diameter) and the electromagnetic field of a Fabry-Pérot optical microcavity (Fig. 1). The cavity has variable length down to 10 microns. It is formed by the ends of two single-mode optical fibres having laser-machined concave faces covered with highly reflecting dielectric coatings. The cavity is pumped, via the fibre, by a laser whose wavelength can be tuned between 760 nm and 820 nm. The nanowire inserted between the two mirrors perturbs the cavity optical field by modifying the optical path, and therefore the frequency of the optical resonance. This defines a "parametric" coupling, where the cavity resonances depends on the nanowire position. The cavity is then kept actively at resonance with the laser by adjusting its length with a piezoelectric actuator.

Fig. 1



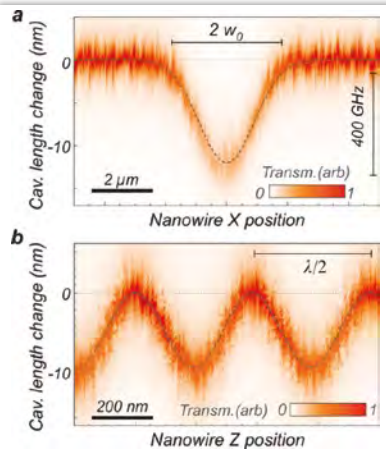
Sketch of and (at right) an actual optical image of the silicon carbide nanowire-oscillator (NW) inserted in the fibre-based optical cavity.

Fig. 2 illustrates the shift of the frequency of a cavity resonance, expressed as a change of the cavity's effective-length, observed as the nanowire is scanned across the cavity both (a) perpendicularly and (b) parallel to the cavity axis *z*. The nanowire acts here as a sub-wavelength sized probe of the cavity optical field and the field's main features (its transverse waist w_0 and the longitudinal standing wave structure) can be precisely characterised. Moreover the dispersive optomechanical coupling can be quantified and amounts to 3 GHz per nm of the nanowire's displacement.

From this experimental value, the intrinsic optomechanical coupling produced by the nanowire's zero point fluctuations can be computed and amounts to $g_0/2\pi$= 1.2 MHz, where g_0 is the optomechanical coupling per photon. That is, a single phonon populating the first flexural mode of the mechanical oscillator would result in a 1.2 MHz frequency shift of the cavity mode. This shift would be 25 times larger than the mechanical resonance frequency (~50 kHz), thus placing the system in the largely unexplored "ultrastrong" coupling" regime. The interaction becomes so extreme that a single photon injected in the optical cavity would exert a radiation force on the nanowire displacing its rest position by more than its zero point fluctuations.

The nanowire is an exquisite force sensor, which has allowed us to explore the optomechanical interaction in the opposite direction and measure the force exerted on the nanowire by the intracavity light field (the "back-action"). Using a second, independent, probe laser, the force exerted on the nanowire by the optical field was measured as the nanowire was scanned along the optical mode of the cavity, allowing us to map the opto-mechanical interaction vector field.
From the above measurements, done at room temperature, we can estimate that the presence of a single photon in the cavity mode would apply an optical force sufficient to displace the nanowire by an amplitude far larger than the spatial extension of its thermal noise, if it were thermalized at very low temperature (20 mK). In turn, the cavity would become bi-stable with just a single photon in the cavity mode. This opens new perspectives in quantum optics, and in particular the investigation of the hitherto unexplored regime of single photon cavity opto-mechanics.

Fig. 2



Relative shift of the cavity resonance (expressed as an equivalent length of the cavity in nm) measured as the nanowire is scanned (a) across the waist (w_0) and (b) along the optical mode

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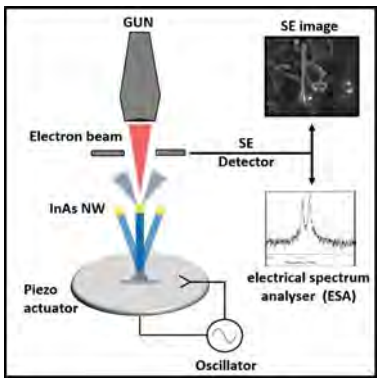
Dynamic Electron Microscopy of nanomechanical resonators

Electron beam microscopy is an important instrument in both research and technology, due to its ability to image matter at the nanometre scale. At the Institut Néel we have been employing a Scanning Electron Microscope (SEM) to observe the mechanical vibrations of nanometre-scale mechanical devices, specifically semiconductor "nanowires". These vibrations are very sensitive to *e.g.* their mass, strain and temperature. This measurement technique has a weak backaction, due to the recoil of each electron hitting the nanowire, which is detectable in our experiments. Proportionately, the recoil of the vibrating target is comparable to that of a basketball player colliding with a fly!

"Nanomechanical" devices are becoming more and more attractive as objects for research and for technological applications. Due to their reduced size and mass, these objects present original and very sensitive properties exploitable in a variety of contexts, such as quantum physics, ultra-sensitive force measurements and mass spectroscopy.

A Scanning Electron Microscope (SEM) builds an image by scanning a focussed electron beam across the target object. Secondary electrons, emitted by atoms excited by the electron beam, are detected with the microscope's secondary electron (SE) detector (Fig. 1). The 1 nm diameter focal spot of the electron beam allows high resolution analysis of nanomechanical devices.

Fig. 1



Scanning Electron Microscope setup for studying nanomechanical oscillators. "NW" = vibrating nanowire. "SE" = Secondary electrons.

At the Institut Néel, we have equipped our SEM installation with a high frequency piezo-electric actuator, which is used for the calibration of the nanowire's thermal-motion amplitude. The electron beam energy is easily tuneable from 100 V to 30 kV and the beam can be readily focussed precisely anywhere on a nanometre scale object.

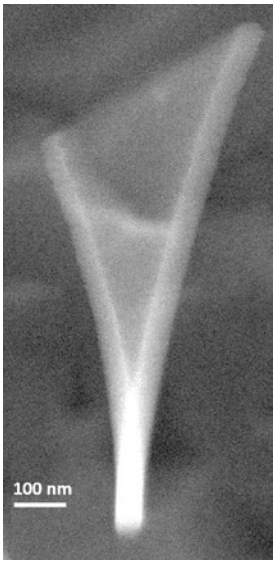
After initial imaging, the electron beam is fixed on an edge of the nanowire. The nanowire's vibrations (thermal or induced) then result in fluctuations of the secondary electron current. The oscillating output from the secondary electron detector is fed to a spectrum analyser (Fig. 1). We have measured resonant frequencies ranging from less than 0.5 MHz up to 15 MHz, depending on the chemical composition and the shape and the size of the vibrating object. Resonance doublets are systematically observed, corresponding to the two vibrational directions of the not-exactly cylindrical nanowires.

We summarize briefly here a study done in collaboration with P. Verlot (Nottingham University) concerning Indium Arsenide (InAs) nanowires grown vertically on an indium phosphate substrate. They had diameters around 70 nm and lengths 4 or 5 microns. We measured and analysed the thermal (Brownian) motion of the InAs nanowires and we demonstrated the existence of a radial backaction process originating from the momentum exchange between the electron beam and these ultralight nanomechanical devices.

During this work, we elucidated some phenomena associated with the electron microscopy technique itself. The mechanical resonance frequencies are very sensitive to changes of the nanoresonator's mass. Continuously monitoring the nanowires' vibrations we observed a significant increase of their oscillator frequency with time. We could fit this behaviour accurately by a model including both desorption and etching. Our nanomechanical devices generally show adsorbed organic molecules at their surface. The electron beam heats the nanowire, which desorbs these contaminants, reducing its mass, thus increasing the vibrational frequency. But the e-beam can also have an etching effect, even taking a "bite" out of the nanowire. And, a very slow deposition of carbon appears caused by the cracking of hydrocarbons present both in the residual atmosphere of the microscope's high-vacuum chamber and on the surface of the specimen.

Our work also explains the widespread observations of researchers using Scanning Electron Microscopes to characterise nanowires. They see vibrations of nanowires anchored on a substrate as a blur on their SEM images. Fig. 2 shows this effect, which we have amplified by resonantly driving a nanowire's motion.

Fig. 2



Scanning Electron Microscope image visualizing an InAs nanowire driven at its resonance frequency.

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FURTHER READING...

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Nanoprobes for imaging deep biological tissue

Bio-imaging consists in taking images of biological items, such as cells, blood vessels, etc, with sub-micron resolution. It aims at understanding biological processes, inflammatory responses or abnormal biological activity and, especially, detecting tumours at an early stage. It makes use of introducing, into the tissues, nanometre scale probes which have been "functionalized" *e.g.* by attaching molecules that target specific receptor cells. Thanks to their physical characteristics, these probes contrast with the surrounding biological tissue, allowing visualisation. With the advent of fluorescence microscopes, optical imaging is booming and strategies are developed to answer challenging problems in nanomedecine. There is potential for imaging with much higher resolution than X Ray, Magnetic Resonance or Positron Emission Topography techniques. In this context, new optically active nanoprobes are needed.

Along with a low toxicity and high bio-compatibility, the specifications for fluorescent, optical nanoprobes require that they can be excited efficiently through the "biological transparency window" (the near-infrared wavelength range 650-1400 nm) by two-photon processes. Scaling as the square of the excitation intensity, these processes allow deep-tissues imaging, occurring only at the focal point of the excitation source, thus limiting tissue autofluorescence and thus enabling high contrast.

In addition, there is a need for multi-functional nanoprobes, combining different physical characteristics for versatile bio-imaging and even to develop "theranostics" tools (combining diagnosis and therapy). In this context, we are studying lanthanum iodate nanocrystals for their potential as multi-functional optical nanoprobes.

We have developed a microwave-assisted hydrothermal synthesis process to obtain lanthanum iodate nanocrystals aiming at sizes smaller than 100 nm. Among the different lanthanum iodate polymorphs, the phase α-$La(IO_3)_3$, crystallizing in a monoclinic crystal system, is of particular interest. This is because, due to its non-centrosymmetric crystal structure resulting from the 3D-arrangement of the iodate group, α-$La(IO_3)_3$ presents strong Second Harmonic Generation (SHG) under near-infrared excitation. That is, this material can convert two photons characterized by a frequency ω, into one photon with a frequency 2ω (and half the wavelength).

We have obtained 50-nm size α-$La(IO_3)_3$ nanocrystals (Fig. 1) through the co-precipitation of lanthanum chloride with iodic acid in water, followed by microwave-assisted hydrothermal treatment at 250°C for a few minutes. Thanks to similar ionic radii and identical oxidation state, lanthanide ions such as Erbium Er^{3+} and Ytterbium Yb^{3+} ions can be substituted for Lanthanum La^{3+}. These rare earth ions have excellent fluorescent characteristics. Similar nanocrystal size and morphology are obtained for α-$La_{1-y-x}Yb_yEr_x(IO_3)_3$ solid solutions with y = 0.1 and x varying from 0.005 to 0.02.

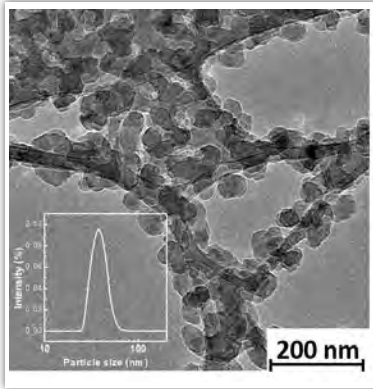

Fig. 1

Transmission Electron Microscope image of α-$La_{0.88}Yb_{0.1}Er_{0.02}(IO_3)_3$ nanocrystals recorded at 77 K. In the inset is shown the size distribution by intensity measured by Dynamic Light Scattering from $La_{0.88}Yb_{0.1}Er_{0.02}(IO_3)_3$ nanocrystals dispersed in ethylene glycol.

We have measured the efficiency of Second Harmonic Generation in these α-$La(IO_3)_3$ nanocrystals, in collaboration with colleagues at the SYMME laboratory (Materials and Mecatronics Lab), Annecy. The SHG efficiency appears to be comparable to that of other well-known second harmonic nanocrystals, such as $BaTiO_3$ and $LiNbO_3$. When doped with the rare earth ions Yb^{3+} and Er^{3+}, our nanocrystals combine both good Second Harmonic Generation properties and photo-induced luminescence by "up-conversion" processes upon near-infrared excitation, *i.e.* in the transparency window of the biological tissues (Fig. 2). Up-conversion means that a first photon excites a rare earth ion to a long-lived intermediate state, and a second photon excites the ion from this state to a high luminescent state.

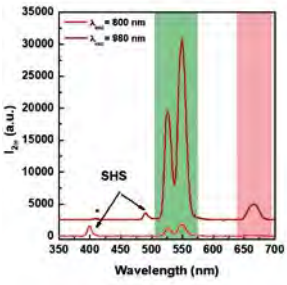

Fig. 2

Non linear optical response of α-$La_{0.88}Yb_{0.1}Er_{0.02}(IO_3)_3$ nanocrystal dispersions in ethylene glycol under a 800-nm excitation (lower curve) and 980-nm excitation (upper curve). For clarity, the spectra have been stacked. The arrows SHS point out the Second Harmonic light coming from the targeted nanocrystals. The up-conversion luminescence of the Yb^{3+} and Er^{3+} ions is evidenced at 525 nm, 550 nm and 660 nm.

Coupling Second Harmonic Generation and Up Conversion processes within a single multifunctional nanoprobe could bring together the advantages of both mechanisms. This combination of Second Harmonic Generation and photoluminescence leads to multi-functional α-$La(IO_3)_3$ nanoprobes with efficient optical properties.

Further prospects include toxicity evaluation for the iodate nanocrystals, as well as, at this stage, *in vitro* bio-imaging on biological samples, taking advantages of the multi-functionality of Yb^{3+},Er^{3+} doped α-$La(IO_3)_3$ nanocrystals to detect lung cancer cells in the framework of the French -Swiss Inter-regional project "OncoNanoScreen" (2019-2021).

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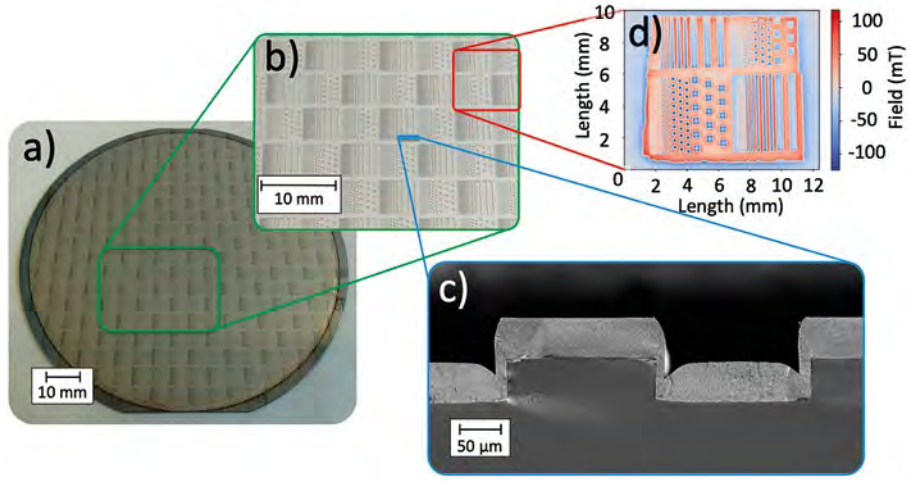
Small magnets with a big future

The discovery of high performance Neodymium Iron Boron (NdFeB) magnets, almost 40 years ago, revolutionised the design of electromagnetic motors, generators and actuators. Today they are key components of a range of devices including gearless wind turbines, electric and hybrid-electric vehicles, electric bicycles, hard disk drives and air-conditioners. At the Institut Néel we have mastered the fabrication of high performance NdFeB "micro-magnets", unlocking enormous potential for the development of Micro-Electro-Mechanical Systems (MEMS) and other micro-scale devices, with applications in areas as diverse as telecommunications, bio-technology and the Internet of Things.

Fig. 1



(a, b) Plane-view optical images of a 50 µm thick NdFeB film deposited on a pre-patterned 100 mm Si substrate together with (c) a cross-sectional scanning electron microscope image of a few micro-magnets and (d) a Magneto-Optical image of the stray magnetic field produced by a part of the film.

The defining characteristic of a permanent magnet is its resistance to demagnetisation, as quantified by its coercivity. The strength of a magnet's stray magnetic field (its external field) is proportional to its remanent magnetisation. Both coercivity and remanent magnetisation are extrinsic properties, their upper limits being set by the material's intrinsic magnetic properties. Their actual values are determined by the magnet's internal structure (size, shape and orientation of main-phase grains, grain boundary phases…) and thus how it is fabricated.

The force of interaction between a magnet and another object is proportional to the gradient of its stray (external) magnetic field. As we scale-down the size of a magnet, while preserving its extrinsic magnetic properties we scale-up its stray magnetic field gradient. As a result, the magnetic field gradient force per unit volume can be increased by orders of magnitude in micro-scale systems compared to macro-scale systems.

Down-scaling the size of NdFeB magnets is not trivial. Machining of sintered bulk magnets (to a lower limit of a few hundred µm) leads to surface degradation, resulting in a loss of coercivity. Besides, it leads to materials waste, and the manipulation and integration of small free-standing magnets is challenging.

We use high-rate triode sputtering to produce high coercivity and high remanence NdFeB films of thickness up to 50 µm, on silicon wafers of diameter 100 mm (Fig. 1). These films can be patterned using standard clean-room facilities, *e.g.* photo-lithographic patterning and deep reactive-ion etching of the Si substrates prior to deposition of the magnetic layer, wet etching, and chemical mechanical planarization. The size, shape and disposition of the micro-magnets can be adapted to suit specific applications. The fact that they are substrate-mounted greatly facilitates integration into complex micro-devices, and the use of large substrates allows for batch fabrication.

Our micro-magnets have already been used in a range of bio-medical applications, in the framework of collaborations with various laboratories. They have also been used to develop prototype micro-mechanical devices with an industrial partner and we are presently involved in ANR (Agence Nationale pour la Recherche) projects to develop thermal and vibrational energy-harvester prototypes, to further demonstrate the enormous potential of our high performance micro-magnets. We have recently started a "maturation" project with the local technology transfer incubator SATT - Linksium (Société d'Accélération du Transfert de Technologies de Grenoble Alpes) to explore the market potential for our micro-magnets and to find new industrial partners who will be clients of our future start-up.

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- *https://www.linksium.fr/en/projet/micromagfab/*

Ridge waveguides for optical frequency conversion

In a non-linear crystal, the induced polarisation *P* of the crystal responds non-linearly to the electric field *E* of an incoming light beam. Such crystals also present birefringence, so a monochromatic light beam experiences different refractive indices, depending on its propagation direction and its polarisation. The birefringence can be exploited to generate new optical frequencies different from the frequency of incoming light. As an example, in second-harmonic generation, two incoming photons of the same frequency and incident on the crystal can give birth to a third photon twice their frequency and half the wavelength, if conditions for phase matching are reached. Thus by pumping a non-linear crystal with an available infrared laser source, one can efficiently generate wavelengths in the visible spectrum.

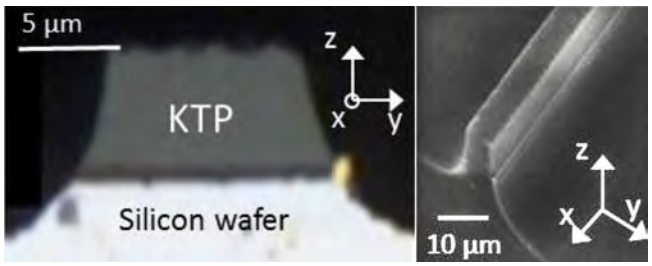
One of the most used crystals in non-linear optics is "KTP" (potassium titanyl phosphate or $KTiOPO_4$). In a KTP crystal, the optimum wavelength for efficient second-harmonic generation can be tuned by using different crystal orientations. In order to combine the properties of KTP with strong confinement of light in view of integrated photonics applications, we are developing KTP optical ridge waveguides having micron-sized transverse dimensions. These waveguides combine high refractive index contrast with stable guiding structures.
However, the effective refractive indices of the crystal are modified by the very small transverse dimension of the ridge which is comparable to the light wavelength. This implies a change of the phase-matching wavelengths for the input and output waves propagating in the crystal. Therefore, prior to doing experiments, we investigated theoretically the changes in the phase-matching conditions as a function of the transverse dimension of the KTP ridge, for the fundamental and second-harmonic waves propagating along the x-axis of the crystal.

Our calculations were then compared with a second-harmonic generation experiment performed at Institut Néel in a ridge waveguide manufactured in collaboration with Femto Sciences and Technologies Institute, Besançon. The 1-mm-thick hybrid structure was composed of a flat KTP rectangle bonded on a silicon substrate and thinned down by grinding and polishing techniques to obtain a KTP layer a few microns thick. The ridge waveguide in Fig. 1, of length L=15.8 mm and transverse section close to S = 38 µm^2, was shaped using a precision diamond saw. This waveguide was tested by pumping it with picosecond pulses from an optical parametric amplifier that is tunable in the infrared region. Fig. 2 gives the second-harmonic output intensity as a function of the input fundamental wavelength. A maximum was obtained for an experimental wavelength close to theoretical expectation, which fully validated our calculations.

Our theoretical prediction of the second-harmonic conversion efficiency, defined by the ratio between the intensities of the fundamental and second-harmonic wavelengths, also fully matched the experimental results, without any fitting parameters. In this first generation of these ridge waveguides, a maximal efficiency of 4.5% has been achieved. Using KTP ridge waveguides with lower losses and extending the waveguide length could lead to even better results, opening a new field for integration of KTP waveguides in photonic nonlinear devices. The corresponding experiments are in progress.

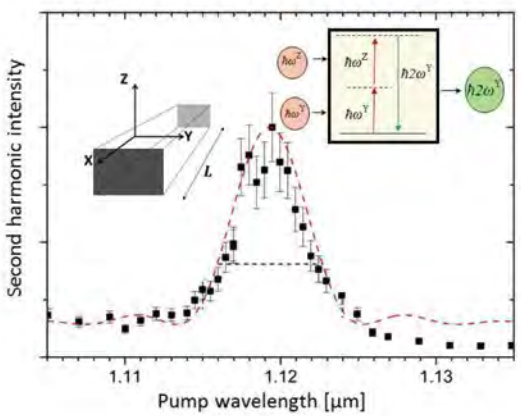
Such very compact photonics device components have potential applications in varied areas, such as medecine (fibroscopy...), biochemical detection, displays, environmental monitoring.

Fig. 1



At left: image of the input section of a KTP ridge waveguide. The crystal is bonded on a silicon wafer. At right: Top view of a ridge waveguide.

Fig. 2



Intensity profile of the yellow-green light beam generated by the waveguide at the second harmonic wavelength as a function of the input infrared pump's wavelength. Phase-matching is achieved for a fundamental wavelength of 1.12 µm, close to the results of the theoretical calculations. Upper-right inset: ħω is the energy of the fundamental photon, ħ2ω the energy of the second-harmonic photon. Superscripts Y and Z are the photons' polarisation directions, perpendicular to the ridge geometry shown at left.

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The "123 Couleurs" project

The Institut Néel has many activities in the popularisation of science, especially working with schools but also outreaching to the general public. In a recent initiative, over the past four years, we have created a science-popularisation website entitled "123couleurs.fr" which is dedicated to the theme of colour. On this website, we propose original pages providing experimental protocols and scientific explanations of many phenomena involving colour. In addition, a commercial activity has been developed for supplying specialized material and pedagogical experiments kits.

The main objective of the project "123 Couleurs" is to create resources for the public (schools, associations, individuals), and to allow them to acquire, by applying a scientific approach and by doing simple experiments, an understanding of phenomena involving colour (*e.g.* mixing and decomposition of light, colour vision, the RGB colour code,…). Colour is a fascinating multi-disciplinary subject, which calls on physics, chemistry, biology and technology. It is also tied to historical and cultural issues, which makes it attractive for both the scientific and non-scientific public. Since 2015, we have created two main on-line activities:

First is our website 123couleurs.fr, which offers three different entry points for visitors, featuring questions (*e.g.* "how many colours are in the rainbow?"), experiments (*e.g.* "colour reflections on a CD surface") and explanations (*e.g.* "the spectrum of light"). We publish approximately one new page every month and, today, three-quarters of the website has been completed. Second is an online store, which sells source materials and equipment difficult to find in the general commerce (colour filters, diffraction gratings, polarizing filters...) and also locally-made pedagogical kits that are available in different forms depending on the target audience (young children, teenagers, school-teachers...).

Three people are involved in the project: Sylvie Zanier from Université Grenoble Alpes, and CNRS researcher Julien Delahaye (who is a member of the Institut Néel's Magnetism and Superconductivity team) are in charge of all scientific aspects of the project, namely the authoring of the website's pages, the selection of the material and the design of the science kits. They both have more than 10 years experience in science popularisation and teaching activities, especially on the theme of colour. Céline Cardeilhac, a microentrepreneur, is in charge of the commercial aspects of the project.

Beyond the development of our website, we take part in many public events, such as local science festivals. We are also involved in ongoing training of primary school and college teachers, via the Maison pour la Science en Alpes Dauphiné.

Fig. 1

Children looking at a flashlight with diffraction glasses. ("Remue-méninges" festival in Echirolles, Isère, France).

In that regard, we have created a kit specially designed for primary school teachers who wish to conduct experiments in their classrooms. One such kit is currently on loan in the Grenoble public-education Académie, for borrowing by regional schools. We also write articles for the high school physics-teacher community and we contribute to society-based issues related to colour, such as the current debate on blue-light hazards.

Fig. 2

At left: The "123 Couleurs" project featured on the front cover of the Bup (Bulletin de l'Union des Physiciens).
At right: An investigation into blue light by the consumer protection association "Que Choisir", to which we contributed.

Our website attracts a growing audience, with now more than 15000 visitors every month. We deliver material and kits all over France and to a wide range of users (primary schools, high schools, universities, museums, associations, individuals). The commercial activity was in balance in 2018 and should be consolidated in the future in order to guarantee the long-term viability of the project.

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HIGH LIGHTS n°13

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