Highlights 2011



EDITORIAL

Institut NEEL is a fundamental research laboratory devoted mainly to condensed matter physics as clearly illustrated by the content of this collection. The activities of the Institut NEEL research groups cover a broad scientific field mainly encompassing quantum fluids, new materials, crystallography, surface science, quantum nano-electronics, nano-mechanics, quantum and non-linear optics, spintronics, magnetism, superconductivity... Our research activities have a significant and growing interface with biology, as illustrated here, as well as with chemistry and engineering sciences.

The large diversity of our scientific activities cannot be summarized in a few pages. However, focusing on thirteen remarkable highlights, we illustrate our commitment to fundamental research, including initiatives in "high risk" directions where success is never guaranteed. Moreover, it shows that through its technology platforms, our laboratory provides our various research teams with the required design tools, fabrication facilities and analysis equipment in order to produce high-level scientific results.

A century after the discovery of superconductivity by Kamerlingh Onnes in Leiden, the commemorations of this anniversary have shown that this research field is still very active in 2011. We were saddened to hear of the death this year of Professor Albert Lacaze, a pioneer in low temperature research in Grenoble who built the first hydrogen liquefier 60 years ago.

The year 2012 will mark the completion of the new building designed to minimize electrical, acoustic, thermal and magnetic interference as well as humidity. Its unique features will enable the laboratory research teams to remain at the highest international level in areas as diverse as quantum information, crystal growth, microscopy, optics and nanofabrication.

Alain SCHUHL Director of Institut Néel, CNRS-UJF

CONTENTS

Supramolecular spin valves	3
Bulk PPKTP by crystal growth from high temperature solution	4
Structural and magnetic chiralities in a langasite compound	5
A bright single-photon source based on a photonic nanowire	б
Quantum melting of a spin-ice as a new route to supersolidity	7
First scientific results of Planck surveyor	8
Fluctuating charge order destroys metallic behavior	10
Nanostructuring surfaces : deconstruction of the Pt(110)-(1x2) surface by $C_{_{60}}$	11
Exploring the quantum world and its new paradigms with neutrons : entanglement and Bose Einstein condensate	12
Surface plasmon polariton scanning quantum source	13
Blocking heat transfer at the nanoscale	14
Emergent magnetic charge crystal in artificial dipolar spin ice	15
Energy minimisation in biological cells	16

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Supramolecular spin valves

The field of Single-Molecule Magnets (SMM) is very promising, since an individual magnetic molecule represents the ultimate size limit for storing and processing information. Magnetic molecules are considered very promising for spintronics – electronics that exploits the spin as well as the charge of the electron – because they can store one bit of information in an extremely small volume. However, in order to use magnetic molecules, one has to find a way to measure their magnetization. Here we show how magnetic molecules could act as building blocks for the design of spintronic devices. A collaboration between experimental chemists and physicists has led to a procedure that combines bottom-up processing techniques with conventional top-down nanofabrication. We have built a novel spin-valve device in which a non-magnetic molecular quantum dot, consisting of a Single-Wall Carbon Nanotube contacted with non-magnetic electrodes, is laterally coupled via supramolecular interactions to a TbPc2 molecular magnet. The localized magnetic moment of the SMM leads to a magnetic field-dependent modulation of the conductance in the nanotube with magnetoresistance ratios of up to 300% below 1 K. Our results open up prospects for new spintronic devices with quantum properties.

A standard Giant Magnetoresistance (GMR) spin valve is an electronic device in which two conducting magnetic layers are separated by a non-magnetic layer. A large modification of the electrical conductance through the device can be achieved by switching the magnetic configuration of the two electrodes between parallel and antiparallel alignments. Under increase of an external magnetic field, because the two layers have different magnetic coercivities, the magnetization of one layer switches at a different field value than the other. That is, the configuration goes from parallel to antiparallel, and finally back to parallel alignment, thus switching the conductance of the device. The resulting Magnetoresistance Ratio is defined by MR = $(G_p - G_{AP})/G_{AP}$ where G_p and G_{AP} are the conductances of the spin valve for parallel (P) and antiparallel (AP) alignment. Typical MR values for metallic spin valves lie in the ten percent range at room temperature. A tunnel barrier between the two layers leads to MR values greater than one hundred percent, as used for instance in commercial spin valves in reading heads.

Our experiments represent the realization of a suggestion we made in 2008 to use quantum nanomagnets: if a Single-Molecule Magnet is coupled laterally to a contacted Single Wall Carbon Nanotube (SWCNT), its highly anisotropic magnetic moment should influence the current passing through the nanotube, and thus permit readout of the molecule's magnetic state by standard conductance measurements. This idea has now been validated: we have measured high magnetoresistance ratios in such a "supramolecular" SMM-SWCNT geometry, in which single quantum nanomagnets act as both magnetic polarizer and analyzer (see Figure). The results demonstrate magnetization switching detected electrically for a single quantum magnet.



A spin valve built entirely from soft organic materials rather than the usual inorganics: A lead made of a Single-Wall Carbon Nanotube (black in the Figure) is contacted at each end by non-magnetic electrodes (gold colour). It is coupled laterally to molecular, TbPc_2 (Pc = phthalocyanine) quantum magnets (green), the latter acting as localized magnetic moments. Sweeping a magnetic field modulates the conductance through the nanotube, achieving magnetoresistance ratios up to 300% between fully polarized and non-polarized molecular configurations.

We have also built a similar device using a graphene nanoconstriction decorated with TbPc_2 magnetic molecules. In this case, a magnetoconductivity signal as high as 20% is found for the spin reversal, revealing the uniaxial magnetic anisotropy of the TbPc_2 quantum magnets. The results show the behavior of multiple-field-effect nanotransistors with sensitivity at the single-molecule level.

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GRAPHENE SPINTRONIC DEVICES WIT MOLECULAR NANOMAGNETS

A. Candini, S. Klyatskaya, M. Ruben, W. Wernsdorfer, and M. Affronte, Nano Lett. 11 , 2634 (2011).

Bulk PPKTP by crystal growth from high temperature solution

Periodically-poled ferroelectric crystals are the nonlinear optical media of the future where the frequency conversion process can be tailored by a periodic structure of ferroelectric domains of alternating polarity, imprinted into the material. These crystals show unprecedented efficiency and other properties that were previously unattainable. However, the sample thicknesses obtained so far have limited their use to low and medium power applications only. We have addressed this problem with a novel growth method that yields larger crystals.

Until now the common method for obtaining a periodicallypoled ferroelectric crystal was the electric-field poling technique. That is, local inversions of the spontaneous polarization are produced in a single-domain crystal using electric fields applied via patterned electrodes on the crystal surface. With this technique the grating periodicity can be controlled very well by lithographic patterning, but the high fields required limit the thickness obtainable to a few millimetres. Various growth techniques have been proposed in order to obtain larger samples with periodic domain structures, but unfortunately the obtained grating structures were not regular enough for use in practical nonlinear devices.

Figure 1 : Flux growth process used to grow large, periodically poled KTP crystals starting from a periodically poled KTP seed.



With the aim of increasing the size of periodically-domainstructured crystals having a controlled and regular grating period, we proposed an epitaxial growth process using thin seed plates that had been domain-engineered by electric field poling. We have demonstrated this process by growing PPKTP, that is periodically poled crystals of the orthorhombic *mm2* ferroelectric KTiOPO₄. The process is shown in Figure 1.

Epitaxial growth of PPKTP was performed onto the two c faces of PPKTP seed plates which had previously been inverted periodically with a grating period of $\Lambda = 38.86 \,\mu\text{m}$ by application of electric fields.

We chose a flux composition that allowed crystal growth below the Curie temperature of KTP and below the roughening temperature of the c faces. Domain-structured KTP layers of up to 800 µm thickness were grown, by a procedure which could be scaled to much larger sizes. A sample is shown in Fig. 2(a). By scanning electron microscopy (SEM) on the *b* face, we could verify that the interfaces were of very high quality and that the domains had propagated through the full thickness of the sample without any measurable variation in size, see Fig. 2(b).

The high optical quality of the grown layer, in terms of regularity and extension of the domain grating, was verified by quasi-phase-matched second harmonic generation measurements using a laser beam propagating along the a axis and probing the $d_{_{33}}$ nonlinear coefficient of the crystal.



Figure 2 : Periodically domain-structured-KTP films grown on a PPKTP seed in a 0.1 KTP – 0.6 KPO_3 – 0.3 KF flux. a) Photograph of a PPKTP crystal and, at left, a cross-section showing the growth seed and its crystallographic axes. b) Scanning Electron Microscope image of the PPKTP seed and the grown layer.

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Structural and magnetic chiralities in a langasite compound

A novel, doubly-chiral magnetic order is found in the structurally-chiral langasite compound $Ba_3NbFe_3Si_2O_{14}$. Our neutron scattering experiments show that the magnetic ground state exhibits a unique sense of rotation of the spins along helices, as well as a unique sense of rotation of the spins around small triangular units in the plane perpendicular to the helices. The spin-wave excitations emerging from this totally-chiral magnetic order present fully chiral spin-correlations over the whole energy spectrum.

Ubiquitous in nature, chirality (from the Greek word $\chi \epsilon_{i\rho}$ for "hand") is the geometric property of an object according to which it exists in two distinct states called enantiomorphs that are images of each other in a mirror. Hands, sea-snail shells, biological molecules like amino acids, even the electroweak interaction, are chiral. In magnetism, an example of chirality is found in helical ordering, where it is related to the sense of rotation of the magnetic moments around a helix axis. Another example of magnetic chirality is the clockwise/anticlockwise sense of rotation of three spins having 120° relative orientations, each situated at the summit of a triangle (the usual signature of "magnetic frustration"). Usually, in materials presenting such spin arrangements, two domains of opposite chirality coexist and are equally populated, yielding a state of zero macroscopic chirality.

A novel chiral magnetic order is found in the langasite compound Ba,NbFe,Si,O,,, single crystals of which are "enantiopure", that is they grow in an atomic arrangement that always shows a single macroscopic structural chirality. Unpolarized neutron diffraction has shown that the magnetic moments are distributed in a 120° arrangement over small triangle units whose centres form a triangular lattice. This order is helically modulated in the direction perpendicular to the triangles. Techniques based on polarized neutron diffraction have proved that a single triangular magnetic chirality together with a single helical magnetic chirality is stabilized in an enantiopure Ba₃NbFe₃Si₂O₁₄ single crystal. We have analyzed the relation between the structural chirality, the 120° spin arrangement, the formation of the helices, and the selection of the magnetic chiralities. It is expected that such totally chiral magnetic states should optimize the potential magnetoelectric and multiferroic behaviours.



Figure caption: (a) helical chirality states (b): Triangular chirality states (c): 120° planar spin structure propagating helically in the perpendicular direction (d); Chiral inelastic scattering measured with polarized neutrons revealing the full chirality of one spin wave branch.

This unique chiral (magnetic and structural) order generates spin waves (elementary excitations) with remarkable properties. Spin waves correspond to coherent motion in space and time of small out-of-equilibrium spin components. In Ba₃NbFe₃Si₂O₁₄, they are visualized in the reciprocal space by two branches rising at finite energy from the elastic Bragg positions characteristic of the magnetic order. Most remarkably, the spin wave branch that corresponds to the Fourier Transform of planar spinspin correlations is found to be totally chiral. This is the first observation of chiral dynamics over the whole energy range of the spin-wave excitations in the absence of an external magnetic field. It highlights how the static chiral properties are extended to the dynamics.

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PARITY BROKEN CHIRAL SPIN DYNA

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A bright single-photon source based on a photonic nanowire

The realization of an efficient, single-photon source - that is a device that can produce photons one by one on demand - is an important goal for quantum cryptography and more generally for the future development of photonic quantum information processing. In this context, semiconductor quantum dots (QDs) are very attractive: at low temperature, they can offer a stable single-photon emission with a nearly perfect radiative yield. However, they are generally embedded in a high index semiconductor matrix that prevents the efficient collection of light in the far field. We have overcome this limitation and demonstrated a very bright single-photon source by inserting the quantum dot inside a novel, well controlled electromagnetic environment: a photonic nanowire.

A photonic wire is a monomode optical waveguide that is made of a high index dielectric material. Specifically, we consider here a structure defined in Group III-Arsenide semiconductors which is shown in Fig. 1. The wire is made of GaAs (refractive index n=3.5) and is surrounded by air (n=1). It contains an InAs quantum dot located near the wire axis, with a free space emission wavelength in the near infrared, around 920 nm. The large refractive index contrast between the wire and air has two important consequences. First, the guided mode can be confined very tightly inside a wire having a 200 nm diameter, which guarantees a good coupling to the emitter. In addition, the coupling to the continuum of non-guided modes is strongly inhibited, thanks to a pronounced dielectric screening effect. As a consequence, the spontaneous emission of the QD is nearly completely funnelled into the guided mode. Next, one has to collect the guided photons efficiently with a microscope objective located above the wire. For this goal, the two ends of the wire are carefully engineered. The photons emitted downward are reflected back into the guided mode by an integrated mirror, made of gold and silica. The upper end of the wire features a conical tip, designed to progressively "deconfine" the guided mode into the air, in order to obtain a more directive far-field emission pattern.

The realization of the device starts with the growth, by Molecular Beam Epitaxy, of a planar structure consisting of an array of self-assembled InAs QDs buried in a GaAs layer. After deposition of the SiO_2 -Au mirror, the sample is "flip-chip" glued on a host substrate and the growth wafer is removed. Finally, photonic nanowires are defined in a top-down approach, using electron beam lithography and a carefully optimized Reactive Ion Etching step.

The performance of the source was investigated in a microphotoluminescence setup, with the source at liquid helium temperature. A pulsed laser beam injects electron-hole pairs that are trapped by the QD. The spectrally-filtered fundamental optical transition, which corresponds to the recombination of a single electron-hole pair, then emits a single infrared photon. The source efficiency, defined as the probability to emit a photon into the collecting cone of the microscope objective after an excitation pulse, reaches a maximum when the emitter is saturated. In these conditions, a record value of 72% was obtained. Simultaneously, intensity correlation measurements have provided the unambiguous signature of a very pure emission of single photons.

Beyond a significant progress with respect to the state of the art, this non-resonant approach opens new perspectives for quantum light sources. In particular, it could be applied to spectrally-broad single-photon emitters, to improve the efficiency of room-temperature single-photon sources. A broadband collection will also be a key point for realizing an efficient source of polarization-entangled photon pairs, using the radiative cascade of a quantum dot.

This work was performed by the Equipe mixte CNRS-CEA "Nanophysique et Semiconducteurs".



Scanning Electron Microscope view of a tapered GaAs photonic wire, supported by a gold mirror (250 nm-thick) and a silica spacer (11 nm-thick). The wire contains a single photon emitter: an InAs quantum dot (seen in the Transmission Electron Microscopy view at the right). The dot is located 80 nm above the silica layer.

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A HIGHLY EFFICIENT SINGLE-PHOTON SOURCE BASED ON A QUANTUM DOT IN A PHOTONIC NANOWIRE"

J. Claudon, J. Bleuse, N. S. Malik, M. Bazin, P. Jaffrennou, N. Gregersen, C. Sauvan, P. Lalanne, and J.-M. Gérard Nature Photonics 4, 174-177 (2010).

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Quantum Melting of a Spin-Ice as a New Route to Supersolidity

Predicted forty years ago, supersolids have received renewed interest recently after a possible observation in ⁴He. While the existence of supersolids is still controversial on the experimental side, numerous examples of such phases have been identified theoretically. To investigate a new route to such exotic quantum phases, we have made a theoretical study of the strong quantum fluctuation effects in a special kind of polarized antiferromagnet that has spin-ice properties at the classical limit.

A supersolid phase is characterized by the coexistence of two contradictory order parameters, a solid crystalline order and a superfluid density (or an in-plane spin order in the case of spin systems). This reflects the spontaneous breaking of two independent symmetries, translation and U(1) rotational gauge symmetry. This simultaneous breaking of two unrelated symmetries is a striking feature and does not have a classical counterpart. In some sense, one could think of the supersolid as a quantum phase which is both solid and liquid. It is thus important to obtain a deep understanding of the conditions and mechanisms for supersolidity and to propose new routes to obtain it.

Certain half-polarized antiferromagnetic spinels can be described as anisotropic magnets on a pyrochlore lattice. In the simplified model case of a spin 1/2 antiferromagnet on a checkerboard lattice (the two-dimensional analogue of a pyrochlore), with only an Ising coupling on the bonds and at moderate external magnetic field, the classical ground state is highly degenerate and follows a so-called "ice-rule" constraint: the lowest Ising energy corresponds precisely to one up-spin and three down-spins on every (flattened) tetrahedron (the light blue tetrahedra in Fig. a). Such a classical state is usually referred to as a "spin-ice".

Once in-plane interactions are included, i.e. away from the classical Ising limit, small quantum fluctuations select certain of the ice-rule configurations and a Valence-Bond Crystal (VBC) insulator is stabilized. In our case, this VBC has been shown to be an exotic phase with paired spins resonating on each tetrahedron. Now, at zero temperature, as quantum fluctuations increase more and more, a quantum melting of this insulating phase occurs. Usually, at fixed magnetization, a transition from a Mott-Insulator to a spin liquid is achieved. However, the residual spin-ice nature of the VBC leads to a double quantum transition point with a novel, commensurate supersolid phase (see Fig b).



Figure : a) Spin 1/2 antiferromagnet on the 2D checkerboard lattice. Up-spins are shown as black dots. Under quantum fluctuations, the spin-ice configurations (made up of light blue flattened tetrahedra with one up-spin and three down-spins) are partially destroyed. Some defects appear: tetrahedra with fully polarized spins (dark blue) and with two up-spins and two down-spins (red), and stabilize the commensurate supersolid. b) Artist's view of the Valence Bond Crystal supersolid phase at commensurate magnetization, showing the resonating plaquettes (dark blue) with inhomogeneous spin density (dots) as well as the in-plane spin order (light blue).

The finite entropy of the spin-ice at zero temperature, the magnetic frustration and the strong quantum fluctuations are responsible for the presence of this exotic phase. Thus, spin 1/2 systems with ice-rule constraints constitute a natural and uncharted terrain for searching for supersolidity, both theoretically and experimentally.

This work was performed with F. Trousselet of MPI-FKF, Stuttgart and D. Poilblanc of LPT/Toulouse.

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First Scientific Results of Planck Surveyor

The Planck Surveyor scientific consortium has published its first results after 15 months of successful operation. One of Planck's key technologies, the 100 mK cooler for its high frequency detectors, was invented at the Institut Néel.

Planck Surveyor is one of the key missions of the Cosmic Vision program of the European Space Agency (ESA). Its primary scientific goal is to map the anisotropies of the Cosmic Microwave Background (CMB), with improved sensitivity and angular resolution compared to the two earlier space missions COBE (1992) and WMAP (2003). At small angular scales, the CMB contains invaluable information about the evolution of the universe from the original Big Bang, allowing astrophysicists to test inflationary models of the early universe or to accurately determine the Hubble constant.



The Planck telescope was launched on 14 May 2009 together with the ESA's Herschel telescope. The two spacecraft were inserted at the second Lagrange point of stability, situated about 1.5 million kilometers out from the Earth as it orbits the Sun. The observation phase began in August 2009 and the first scientific results based on two complete sky surveys were presented during an international conference held in Paris in January 2011.

The CMB is basically a blackbody radiation at 2.725 K, a light emitted 13.7 billion years ago, about 380000 years after the Big Bang. The anisotropies under study are small deviations (of order 100 microK) from an ideally uniform blackbody radiation. Planck has two instruments, the low frequency instrument (LFI) and the high frequency instrument (HFI), which cover nine frequency bands between 30 GHz and 1 THz. At these wavelengths, several astronomical sources also contribute to the signal. The main contributions come from cold dust, synchrotron emission and free-free emission from our own galaxy. In order to obtain precise information about the CMB itself, a first step in the data reduction is the "component separation", and the first published results concern the component separation analysis.

Many results are based on the "Early Release Compact Source Catalog", which is a public database containing more than 15000 sources detected by Planck (Fig. 1). The final catalog will be published early in 2013, along with the main cosmological results.

Figure 1 : Position on the sky of all compact sources detected by Planck.



Figure 2 : At left: Map of the Cosmic Infrared Background at 857 GHz. At right: Typical spectrum for a galaxy having redshift 0.6, and the High Frequency Instrument's detection channels most sensitive for such a spectrum.



Figure 3 : One newly detected galaxy supercluster PLCK G334.8-38.0: At left: Planck's Sunyaev Zel'dovich map superimposed with X-Ray contours filtered from the X-Ray image at right, acquired by the XMM-Newton telescope.

Among the recent results, one concerns the Cosmic Infrared Background (CIB). The CIB, which is about 50 times weaker than the CMB, consists of the cumulative infrared emission from all galaxies throughout cosmic history. Being produced by the dust within galaxies, it carries a wealth of information about the processes of star formation. Due to the wide spectral coverage of the Planck instrument, the CIB has been detected at low frequency for the first time (Fig.2).

Another novel result has advanced our knowledge of the star formation process within clumps of cold gas and dust in the Milky Way. Planck has characterized the "anomalous microwave emission", which is a diffuse glow associated with the dense and dusty regions of our galaxy. The new Planck data confirm the theory that the glow is coming from dust grains set spinning at ~1010 Hz by collision with either fast–moving atoms or ultra-violet photons.

A third example concerns the Sunyaev-Zel'dovich effect (SZE), through which distant galaxy clusters imprint a distinctive signature on the CMB. As the CMB photons travel through the Universe, they interact with matter. In a galaxy cluster, the photons scatter off the free electrons present in the hot gas that pervades the cluster. Since the hot electrons are much more energetic than the CMB photons, this interaction scatters the photons to higher energies. Along the line of sight of a galaxy cluster, the CMB photons appear fainter at low frequencies and brighter at high frequencies, with the transition at frequency 217GHz. Combined with observations by the ESA's XMM-Newton X-ray satellite, this gives important information about the galaxy cluster's mass and distance. The Planck collaboration has identified 199 clusters so far (the largest catalogue), including 20 previously unknown clusters. Two of the sources even appear to be "superclusters" (Fig. 3).

Planck's scientific results would not have been possible without the success achieved in challenging development work on cooling systems for very low temperature instruments. Planck's cryogenic chain combines a passive cooling with two cryo-coolers. A closed-cycle sorption cooler uses hydrogen as the working fluid to provide a 20 K stage for both instruments. For the HFI, the second stage is a Joule-Thompson cooler that uses helium to precool to 4K. The complete satellite was launched at room temperature and its cryogenic systems were activated in orbit to reach their final operational temperature after fifty days.

The HFI instrument then uses a novel dilution refrigerator to cool its detectors to 100mK. This system was developed by Air Liquide in Sassenage in collaboration with the Néel Institute at Grenoble and IAS at Orsay. The HFI project was financed by the CNES (the French National Space Agency) which continues to support the development of very low temperature coolers within several R&D programs at the Néel Institute, at the CEA's INAC, and at Air Liquide.

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Planck Early Results series in A&A (Astronomy & Astrophysics) *see also* http://www.esa.int/SPECIALS/Planck

Fluctuating charge order destroys metallic behavior

Phase transitions are ubiquitous: from the crystallization of water into ice, to the alignment of electron spins inside a magnet, to the emergence of superconductivity in a cooled metal. In the case of continuous, second-order, transitions, the transformation from one phase to the other does not come suddenly: it is generally announced by a strong increase in the fluctuations of an "order parameter". The most familiar example is the phenomenon of critical opalescence, reflecting the increase in density fluctuations at the approach of the liquid-gas transition.

In recent years, there has been a growing interest in critical phenomena taking place at (or sufficiently close to) the absolute zero of temperature. This has revealed a new class of phase transitions, called quantum phase transitions. In contrast to ordinary phase transitions, where the control parameter is the temperature, quantum phase transitions are driven by the zero point quantum fluctuations associated with Heisenberg's uncertainty principle. Quantum Critical Points (QCPs) typically occur in physical systems where several ground states are in competition, so that it is possible to tune from one ground state to another by adjusting an external control parameter such as pressure, magnetic field, or chemical composition.



Phase diagram illustrating the emergence of non-Fermi liquid properties induced by the proximity to a charge ordering transition driven by Coulomb electron-electron interactions.

The proximity to a quantum critical point can result in a very unusual state of matter. In a metal, for example, the electrons are so strongly scattered by the quantum critical fluctuations that significant deviations from the usual "Fermiliquid" behaviour can arise, a fact that has measurable consequences -- the analogue of the phenomenon of critical opalescence. More strikingly, the system might even prefer to avoid such anomalous behavior and adopt other forms of order such as superconductivity, novel types of magnetism, or other forms of "hidden" order. Proximity to a quantum critical point may be the cause of anomalous properties in a variety of materials even when the QCP itself is not observed.

While most studies of quantum criticality in recent years have been devoted to the study of QCPs at magnetic phase transitions, QCPs related to charge order instabilities have remained mostly unexplored. In the present work, we have focused on a theoretical model that describes the charge ordering driven by the strong Coulomb repulsion between electrons, as observed in a class of layered organic conductors -- the quarter-filled theta-(BEDT-TTF)₂X salts. Upon applying pressure or by chemical substitution, the critical temperature Tc of the transition can be made to vanish, and a quantum critical point is obtained.

Our theoretical results show that the properties of the electronic system are strongly affected by the existence of the QCP. A new temperature scale T* emerges in the normal phase, above which the elementary electronic quasi-particles that are at the very basis of the Fermi liquid behaviour are strongly slowed down and eventually disappear [Fig.1]. The resulting state exhibits a "bad" metallic behaviour, testified by a strong increase in the effective mass and a consequent reduction of the system's kinetic energy. At the same time, the "Drude" conduction characteristic of metals is strongly suppressed.

These observations agree quite well with what is observed in the theta-(BEDT-TTF)₂X salts by both transport and optical spectroscopy measurements. The emergence of superconductivity in the theta-(BEDT-TTF)₂I₃ compound -the only superconducting member of this class of materials, which is also the one lying closest to the quantum critical point – could well be related with the present scenario. Also, due to the universality of the concepts involved, phenomena similar to those evinced here should in principle apply also to other classes of materials exhibiting charge order, such as the transition metal dichalcogenides.

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L. Cano-Cortés, J. Merino and S. Fratini, Phys. Rev. Lett. 105, 036405 (2010).

Nanostructuring surfaces: Deconstruction of the Pt(110)-(1x2) surface by C_{60}

The interactions at the interface between large molecules and metallic substrates have attracted considerable interest due to the development of new devices based on organic films and/or functionalized inorganic molecules. When deposited on metal surfaces, the C_{60} (fullerene) molecule creates regularly ordered, nanometre scale holes ("nanoholes"). Grazing incidence X-ray diffraction reveals a c(4×4) reconstruction induced by C_{60} on a Platinum (110) surface that had initially a (1×2) type reconstruction. While the initial missing row of the (1x2) surface structure is partially deconstructed, under each fullerene we find a double atomic vacancy involving the topmost Pt layers. The resulting interface is deeply modified with 75% occupancy of Pt and regularly distributed double vacancies. The orientation of the C_{60} molecule is compatible with a *cm* symmetry of the local surface, with one of the molecule's pentagonal rings of carbons almost parallel to the surface and its hexagonal rings almost parallel to the (111) facets of the Pt nanohole.

Two-dimensional ordering of molecules on a surface is often accompanied by a restructuring of the surface involving the two to three topmost atomic layers: such a modified interface may be used for both surface moulding and surface nanopatterning, with relevant technological applications. The mechanism of anchoring of a molecule on the surface is usually quite complex due to an interplay between intermolecular bonds and molecule-substrate interactions involving a large number of adsorption sites. This may result in substrate reconstructions, involving extensive mass transport, which can be accompanied by a structural deformation of the adsorbed molecules.

We studied the $c(4\times4)$ reconstruction obtained by depositing a single layer of fullerene on the Pt(110)-(1×2), missing row reconstruction, using grazing incidence X-ray diffraction at the European Synchrotron Radiation Facility in Grenoble. The substrate was maintained at about 800 K during deposition, and the saturation coverage was determined by monitoring one superstructure reflection during the growth. A set of 45 fractional rods and nine crystal truncation rods were measured to optimize calculation of the structure (Figure 1 shows a few of them).



Figure 1 : X-Ray diffraction intensities measured when rocking the crystal sample in a multiple directions were used to determine the c(2x2) structure of $C_{_{60}}$ molecules on Pt(110). Data shown here are for some fractional rods and crystal truncation rods. The continuous lines correspond to the best fit model. The X-Ray Diffraction data analysis agrees with a surface substrate ordering where the initial (1x2) missing row is "de-reconstructed" by a partial filling (50%) of the missing rows, due to the interaction of the C_{60} with the substrate (Fig. 2). The resulting surface consists of a Pt top layer where complete rows alternate with half-filled ones, yielding a 75% occupancy, with double vacancies alternating with Pt doublets which generates the new c(4x4) surface periodicity. The unit cell contains a single bi-atomic Pt-surface hole used to host the C_{60} molecule and forming a regular, dense quasi-hexagonal molecular phase.



The strong interaction of the C_{60} molecules with the Ptsubstrate is shown by the high number of C-Pt bonds participating in the anchoring of the molecules to the substrate. The most stable configuration yields 15 C-Pt bonds with an average bond distance of 2.4 Å, which is an indication of charge delocalization from the molecule to the substrate over a rather large contact interface area.

This work was done in collaboration with the Institut de Ciència de Materials de Barcelona and the Physics Department of the Universitat Autònoma de Barcelona.

Figure 2 : Top and lateral view of the C_{60} /Pt(110)-c(4×4): Darker color atoms correspond to deeper surface atoms.

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Exploring the quantum world and its new paradigms with neutrons: entanglement and Bose Einstein condensate

There is an interesting parallelism between quantum entanglement and the formation of spin-dimers in quasi one dimensional (Quasi-1D) Heisenberg S=1/2 compounds and Quasi-1D S=1 systems. The essence of quantum entanglement lies in the correlations between parts of a composite system such that the corresponding wave function is not just a product of the wave functions of the individual systems. The well known example of a bipartite entangled state is the spin singlet state, $|\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle$, of two spin-1/2 particles which cannot be written as a product of individual spins. As it turns out, this very same spin singlet state is the ground state of the whole class of Quasi-1D, S=1/2 dimers and S=1 compounds, thoroughly studied during the last 30 years. The physical picture (including phase diagrams and possible phase transitions) has been understood within the formalism of solid state physics alone, whereas the possibilities offered by the quantum entanglement aspects have been explored only recently.

The resulting similarity has excited the scientific community and brought Quasi-1D compounds having antiferromagnetically coupled quantum spins (including the so-called spin-liquids) to the forefront of the possible applications of quantum entanglement to information processing. Such applications include quantum teleportation, super dense coding, quantum cryptography, etc. This is even more tantalizing as some of these compounds (mostly copper oxides) may exhibit non-negligible entanglement at room temperature. The question that remains very much open is to determine which macroscopic quantities can be used to detect entanglement, also called "entanglement witnesses", in condensed matter physics. It was initially proposed that the quantity called "concurrence" is a good measure of entanglement. It was later realized that, for a spin-chain system, concurrence can be expressed in terms of pure spin-spin correlation functions.

Recently it was shown that neutron scattering experiments as well as magnetic susceptibility experiments enable one to quantify the quantum entanglement. This is a very important outcome as neutron scattering techniques can be used to directly quantify all sources of bipartite entanglement present in a system. Therefore, and in addition to describing the maximally entangled Bell states on a single site, neutron scattering allows one to characterize the entanglement between distant particles, a variable relevant to quantum information processing. Classical techniques meant to explore bulk matter can be used to explore the kingdom of quantum physics and help with unveiling the new paradigms.

Here we report the direct observation of a Bose Einstein Condensate (BEC) created solely by antiferromagnetically entangled s=1/2 spin pairs in a quasi-one dimensional compound, the spin-ladder cuprate $Sr_{14}Cu_{24}O_{41}$, by Inelastic Neutron Scattering (INS) experiments. The entangled pairs are composite bosons having a spin singlet ground state, and the lowest energy excitations result from the population of the first excited state, a spin triplet, separated from the ground state by an energy gap of magnitude 32.5meV. Below a characteristic temperature, we observe the appearance of a conspicuous peak at the energy of the



Unpolarized neutron energy scan showing the gap of the spin fluctuations of the ladder subsystem at T=5K. This is a rather sharp excitation in momentum Q and in energy and it shows that the state is phase-coherent. The red and blue points represent two different Q-positions; the latter one can be considered as a measure of the background and non-magnetic contributions. The insert shows the ladder structure, with Cu atoms shown in orange and oxygen in light blue.

gap and $q_{_{Q1D}}$ = π , which remains unresolved in energy and momentum along the Quasi-1D direction. We conjecture that all the triplets making up this peak have the same phase, and we therefore interpret it as the signature of the occurrence of quantum coherence along the ladder direction between the entangled spin pairs. We predict that this condensate is bound to play a fundamental role in explaining the onset of unconventional superconductivity. Indeed, the observed peak at 32.5meV shares many features with the resonant peak observed in INS experiments in the high-T_c cuprates.

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Surface plasmon polariton scanning quantum source

A crucial obstacle to the further miniaturization of electrical devices is the difficulty of making electrical contacts between nano-objects. Indeed, energy transfer and communication between devices at the nanometer scale has become a major issue in developing nano-electronic or nano-optical devices. Recently, photonic and plasmonic methods have been proposed to address these problems. Surface Plasmon Polaritons (SPPs), which are hybrid state electron-photon modes propagating at the interface between a metal like gold and an insulator (e.g., glass or air), are naturally adapted for two-dimensional applications. As surface waves, SPPs are exponentially damped in the directions perpendicular to the interfaces and can be guided in structures that are miniaturized beyond the diffraction limit. These appealing properties make

Relevant to this, we recently developed highly stable, scanning single-photon sources that operate at room-temperature. This was achieved by controlled grafting of a selected nanodiamond (size about 20 nm) hosting a single nitrogen impurity-lattice vacancy (N-V) color-center at the apex of an optical probe (a chemically etched fiber tip). A variant of this method is to graft a larger number (typically 3 to 5) of unselected fluorescent nanodiamonds.

We now report the implementation of these nanodiamond tips for launching Surface Plasmon Polaritons on gold films. A nanodiamond-based tip is excited by 488 nm light (which does not itself excite SPPs efficiently in gold) and brought into the close vicinity (20 nm distance) of a thick, nanostructured Au film imaged by Near-field Scanning Optical Microscopy (NSOM). As illustrated in Figure 1, we have demonstrated that SPPs are efficiently launched by the red near-field light emanating from the fluorescent nanodiamonds situated on the fibre tip. When the number of N-V centres is reduced down to two (hosted by a single 20 nm diamond), single SPPs are launched into a nanostructured film. This opens the way to innovative studies in "deterministic" scanning quantum plasmonics.



Deterministic quantum plasmonics. Left: A single diamond nanocrystal containing two NV color centers is glued at the apex of a bare fibre tip. The fluorescence of the color centers is coupled to Surface Plasmon Polaritons propagating along a gold film. Light is subsequently collected by a microscope for either leakage radiation measurements or Near-field Scanning imaging (NSOM). Right: A typical, intensity-time, second-order correlation function of the photon emission from the two color centers at the tip apex, showing the quantum nature of the emitted light. Bottom: A typical near field optical image of a gold nanostructure acquired using such a quantum plasmonic device.

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NEAR-FIELD OPTICAL MICROSCOPY WITH A NANODIAMOND-BASED SINGLE-PHO-TON TIP

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Blocking heat transfer at the nanoscale

Phonons are tiny packets of vibrational energy, quanta of vibration of the lattice. When acoustic (i.e. low frequency) phonons propagate, they give rise to thermal conductance, the property of a solid to conduct heat. At room temperature, many phonons are present and they are strongly scattered by each other as well as by electrons or impurities; the transport is called diffusive. At low temperature, the probability of phonon scattering greatly decreases and, finally, heat transport will be limited only by scattering on the rough surface of the solid. At even lower temperature, the wavelength of phonons becomes so large that the surface behaves like a mirror for them: they undergo specular reflection. In this "ballistic" regime, phonons will conserve their energy until they reach a thermal bath, a large reservoir where they can finally thermalize with their counterparts.

Working in the ballistic regime with silicon dielectric nanowires having cross-sections of order 200 nm, we have found that we can block heat transfer by blocking the circulation of ballistic phonons. Introducing a structured serpentine into a nanowire (see Figure), we found that the thermal conductance was considerably reduced as compared to a straight nanowire having the same length. This effect was detected by very sensitive thermal conductance measurements on the two different types of nanowire. The phonons can be considered as spheres that are reflected by the serpentine and then cannot transport heat towards the thermal reservoir. The phonons are blocked in the centre of the nanowire, between the two serpentines. This nano-engineering trick provides a useful way to improve the performance of thermoelectric components. Such a device can be called an electron transmitting/ phonon blocking system. Because there is nothing of a quantum nature here, the idea can be implemented at a smaller length scale at room temperature, to improve the factor of merit of nano-thermoelectric devices.



The Scanning Electron Microscope photograph shows a serpentine section in a silicon nanowire. The curves represent the thermal conductance of two different types of nanowire. One nanowire is straight and serves as reference. The others are serpentine nanowires, and show a reduced thermal conductance, thus illustrating blocking of heat transfer.

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Emergent magnetic charge crystal in artificial dipolar spin ice

Using nanofabrication processes, we have synthesized a mesoscopic assembly of interacting nanomagnets, with statistical properties quantitatively described by an Ising-like spin Hamiltonian. This artificial realization of a spin model has been imaged in real space using a synchrotron-based magnetic imaging technique. The magnetic configurations we obtain after demagnetizing the spin assembly fit well with predictions from Monte Carlo simulations. In particular, we observe the emergence of a phase where spins fluctuate while classical magnetic charges, associated with these spins, crystallize. This phase was predicted theoretically, but never observed experimentally. Our results open the way to the investigation of a wide variety of classical spin models within the concept of the "lab on a chip".

The property called "frustration" arises when all the pairwise interactions in a system cannot be satisfied at the same time, because of a system's symmetry, geometry or topology. In some cases, frustration effects lead to an extensively degenerate ground state, i.e a low temperature manifold built with a large number of configurations with identical energy. Pauling's description of the lowtemperature proton disorder in water ice was perhaps the first example of frustration in condensed matter physics, and remains the paradigm. In recent years, a new class of magnetic compounds has been characterized in which the disorder of the magnetic moments at low temperatures is analogous to the proton disorder in water ice, hence the name "spin ice".

Recently, this correspondence between water ice and its magnetic equivalents has been pushed even further with the realization of artificial, two dimensional analogs of spin ice models using nanofabrication techniques. Using lithography techniques, we have made geometrically frustrated arrays of nanomagnets on a kagome lattice, i.e. a lattice of triangles sharing their corners on which the nanomagnets are located (see Figure). The magnetic configuration of each nanomagnet is then determined by X-ray PhotoEmission Electron Microscopy (XPEEM). Due to their elongated shape, magnetization within the nanomagnets can only point along the long axis of the elements. These pseudo spins can also be considered as magnetic dipoles, which allows one to map any spin configuration onto a magnetic charge configuration, the charge at each vertex of the honeycomb lattice being the sum of the pole signs (south = -1, north = +1) of each spin of the kagome lattice participating in the vertex.

A major property of the effective spin Hamiltonian is that, at low temperatures, frustration constrains the spin configurations on each triangle of the kagome lattice such that two spins point inward in the triangle and one outward, or two spins point outward and one inward. This property, also called the "ice rule", is equivalent to constraining the magnetic charge on each vertex of the honeycomb lattice to be equal to plus or minus 1. Combining Monte Carlo simulations and XPEEM magnetic imaging of these artificial arrays, we found an important result. Contrary to what was thought until now, the long range dipolar interaction between the nanomagnets can not be neglected, and actually drives the physics we observe. This result has profound consequences: while the main interest for frustrated compounds arises from the



massive degeneracy of their ground state, this degeneracy is fully lifted when long range, dipolar interactions are included in the model.

Understanding whether or not these long range interactions influence the local spin configuration in artificial arrays of nanomagnets is thus essential, especially because these networks are often considered as a playground to study magnetic frustration effects on a mesoscopic scale. To demonstrate this result, we compared predictions from dipolar spin ice models and our experimental observations. In particular, as the system reaches low-energy spin configurations, it goes through a (predicted) phase transition where spins fluctuate while the magnetic charges at the vertex crystallize to form a perfectly ordered pattern of alternating +1 and -1 magnetic charges. The "beauty" of our work was to observe the emergence of the phase in which the magnetic charges crystallize.

This work was done in collaboration with the Jean Lamour Institute in Nancy, and the SOLEIL and ELETTRA synchrotron radiation facilities.

XPEEM magnetic image of an artificial array of nanomagnets with typical dimension 500x80x10 nm³. The direction of the incoming x rays is indicated by the arrow hv. Black and white contrast is then observed according to the sign + or - of the magnetization component along this x ray direction, as sketched in the two panels below the image. The spin state of each nanomagnet, and thus the magnetic charge carried by each vertex, is measured. The partly superimposed dots highlight the nodes of the kagome lattice.

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Energy minimisation in biological cells

From a thermodynamic point of view, biological cells are non-equilibrium, open systems which are very difficult to describe by the methods of physics. The biochemical machinery that regulates cells is very complex and is controlled in part by the interactions of cells with the surrounding proteins. Using microfabrication techniques developed and patented at the Néel Institute, which are suitable for manipulating fragile items such as proteins, we can control the adhesion of cells to a substrate at the micron scale. Experiments using these techniques together with numerical simulations have shown the property of biological cells to minimize their energy.



Reproducible geometric shapes observed when cells adhere on microstructured substrates. The actin cytoskeleton is marked in red, and green points are accessions. The distance between two member points is 10 microns.

Patterns of proteins were used to control cell shape in a surprising and highly reproducible way (see Figure). In statistical studies conducted on groups of cells, the cell shape most likely to be observed was the square shape, the one that respected the symmetry of the protein network. For dynamic objects in motion and in constant reorganization, the observation of a predominant form suggests the existence of general physical laws governing the organization of such elementary living systems. We have performed Monte Carlo simulations to obtain an expression for the energy minimized by the cells. It involves the energies of adhesion, contraction and compressibility of the cytoskeleton, which are very important biological parameters involved in cell adhesion and in the regulation of the cell's biochemistry. Our simulations have established a phase diagram for the shape of cells adherent on a protein pattern. The diagram is based on dimensionless parameters that describe the competition between the cell's capability to contract and its capability to deform, and that take account of the dispersion within a population of cells.

Since a cell's shape is determined by biochemical activity, tracking the changes in the phase diagram that are induced by drugs gives quantitative information on the biological mechanisms of the drugs. Our current work integrates Nano Electrical-Mechanical Systems (NEMS) and microfluidics to study in situ the rheological properties of contractile elements at the subcellular level, for cells whose architecture is controlled using microfabricated patterns.

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