IPCMS News
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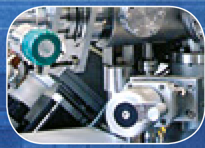

Summary

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Editorial

This new issue of IPCMS News is born with a new direction for our laboratory. Since January 1st 2018, I have the honor to assume the direction of this great institute, together with Rodolfo Jalabert as Deputy Director, and a new management team. In writing my first editorial, I take the opportunity to warmly thank former Director Stefan Haacke, and former Deputy Director Carlo Massobrio for all their valuable work done towards fostering an outstanding environment for research. Their constant commitment allowed us to inherit a vibrant institute, and we expect that they will keep on contributing to the laboratory success through their excellent research.

Paraphrasing a famous Latin expression, directors pass, science remains. This issue reports recent developments that illustrate the wealth of interdisciplinary research that is being conducted in our institute. Strong interaction between physicists and chemists, as well as between experience and theory, result in remarkable achievements in the field of materials and nanoscience. First of all, how doing better at a lower cost? This is the purpose of an exemplary collaboration of M. Boero with Japanese colleagues of University of Kyoto on computer-aid in silico design of catalysts. Turning to an outstanding example of contribution of theory to physics, the results of the team of G. Pupillo bring a valuable insight on vortex glass formation in superconducting systems, beyond the well-established mechanism of pinning by impurities. On the materials side, nanostructures remain at the heart of our concerns. Thus, designing functionalized soft ferrite oxide nanoparticles, organizing them in large assemblies, and controlling their magnetic properties was the focus of another international collaboration involving chemists of the IPCMS (B. Donnio) and of the University of Pennsylvania. Nanoparticles, again, lead to intriguing results when organized on a graphene substrate. J.-F. Dayen and coworkers showed that an assembly of aluminum clusters on a graphene sheet may constitute a single-electron device. These intriguing properties have been observed, in this case, thanks to the development of a particular growth process of such heterostructures. Deepening the knowledge in the physics of quantum systems, is one of the challenges addressed by P. Gilliot's team. Thanks to the use of high quality GaN quantum well and appropriate optical pump-probe setups involving optical coherence decay and spin relaxation measurements, the microscopic mechanism governing spin-dynamics was unraveled and new nanophotonic devices are expected to emerge. Governing properties by design is a chief motivation of G. Rogez and coworkers, using for this purpose organic functional bricks to modify inorganic host structures. In collaboration with French colleagues of Caen and Brest, the IPCMS team succeeded in conceiving new hybrid system exhibiting magneto-electric effect, which was rationalized thanks to exemplary association between chemistry, experimental physics and theory. ■ ■ ■

Université de Strasbourg

Institut de Physique et Chimie des Matériaux de Strasbourg

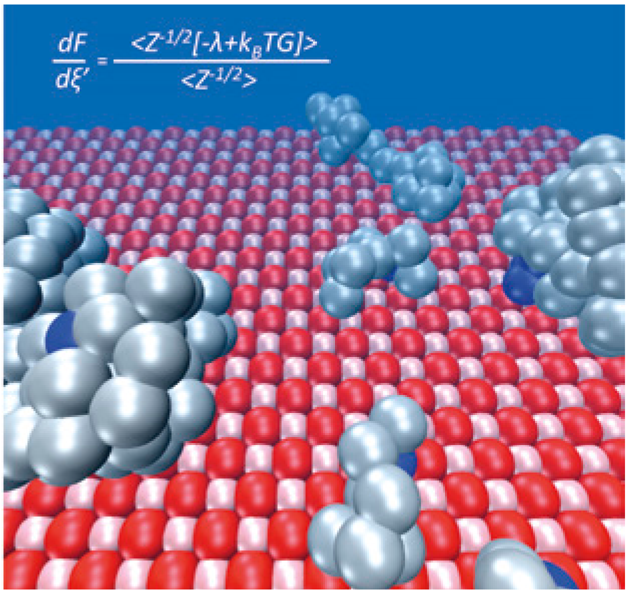
■ ■ ■ Finally, decreasing the spatial dimensions and shortening the time resolution, make it necessary to dispose of tools capable to probe matter and the molecule properties at the atomic scale. Such endeavor was the purpose of G. Schull et al. experiment using scanning tunneling microscope to develop optical spectroscopy at sub-molecular resolution: a real breakthrough for understanding fluorescence of a single molecule and the interaction with its environment.

The IPCMS continues to be strongly involved in international programs. The Indian-French Laboratory for Solid State Chemistry has been renewed. The Brazilian-French Laboratory for Advanced Electron Microscopy of Biomaterials (AEMB) has been launched. The partnership with Korea was strengthened through the visit of a delegation from the university of Ulsan in October 2017 and by our involvement in the new Joint International Laboratory on Building Blocks For Future Electronics (UMI 2B-FUEL) between CNRS, Yonsei university and Ewha university. Last but not least, IPCMS members initiated a new graduate school project on Quantum nanomaterials and nanoscience, and they were successful in a very competitive national call. This new international training program (EUR QMAT) reinforces the position of University of Strasbourg as a center of excellence in the field of quantum technologies.

Pierre Rabu, *Director*

◆ *Downsizing platinum catalysts to minimize the cost and maximize the conversion of toxic air into a breathable one*

Platinum is widely used as a three-way catalyst and considered as an effective material to convert toxic gases into harmless ones. As such, it has a wealth of applications from fundamental catalysis to automobile industry (see e.g. *Nature* **2007**, 450, 334-335), yet its limited availability and consequent high cost represent the two major stumbling blocks to its use. Our dynamical simulations enhanced by a free-energy sampling demonstrated the feasibility of the inhibition of the parasite process knowing as "sintering" of platinum clusters on metal oxides. Specifically, nickel can act as an anchor of Pt nanoparticles (NPs) on the substrate. These results pave a viable route to produce cost-effective and sintering-preventing Pt-based NPs without spoiling their catalytic efficiency, keeping the amount of precious metal to a minimum (nano)amount, and contributing to reduce the problems of air pollution. Our work is also an example of computer aided *in silico* design of catalysts. ■



K. Koizumi, K. Nobusada, M. Boero,
Chemistry Eur. J., **2017**, 23, 1531-1538 – *frontispiece issue.*
DOI: 10.1002/chem.201604188

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IPCMS News

◆ *Intrinsic Disorder in Monodispersed Vortex Ensembles*

The vortex glass is one of the key states in the theory of magnetic and transport properties of type-2 superconductors in the presence of disorder. Within this frame, the glassy phase is caused by the pinning of vortices by impurities and is absent in a clean sample. In the work recently published in Physical Review Letters, we demonstrate that a vortex glass state can be an inherent property of a superconducting system characterized by multiple coherence lengths. In a more general context, our work demonstrates that a structurally disordered glass state of matter can be obtained in the absence of disordered substrates for a simple two-dimensional monodisperse ensemble of particles interacting via isotropic, repulsive, ultrasoft interactions. This is surprising as those conditions are usually associated with minimal frustration. The glass phase appears below a non-equilibrium glass transition temperature and extends to the lowest temperatures examined. Building upon previous results on clustering with ultrasoft potentials, in our work, we provide a description of the microscopic mechanism responsible for the appearance of glassiness in terms of an effective polydispersity that emerges following a quench due to multi-scale interactions. In typical glass forming liquids, frustration results from polydisperse mixtures of particles. In the present systems, the appearance of glassiness stems instead from the effective polydispersity of clusters sizes.

For what concerns experimental realizations, several works have recently discussed "type-1.5 superconductors" that are characterized by multiple coherence lengths, some of which are larger and some smaller than the magnetic field penetration length. These multiple coherence lengths arise in superconducting states that break multiple symmetries and also in materials with multiple superconducting bands. Multiple repulsive length scales can be obtained in thin films of type-1.5 materials due to stray fields or even in artificially fabricated superconducting bilayers, where the different layers give rise to two coherence lengths and the shape of the effective vortex-vortex interactions can be effectively engineered. Due to the generality of the underlying mechanism for glass formation, we expect that the present results may find applications in the context of the physics of ultrasoft particles in fields as diverse as classical colloidal physics and ultracold quantum gases, where similar interactions potentials are obtained, in addition to vortex matter. ■

R. Diaz-Mendez, F. Mezzacapo,
W. Lechner, F. Cinti, E. Babaev, G. Pupillo,
Phys. Rev. Lett. **2017**, 118, 067001.
DOI: doi.org/10.1103/
PhysRevLett.118.067001

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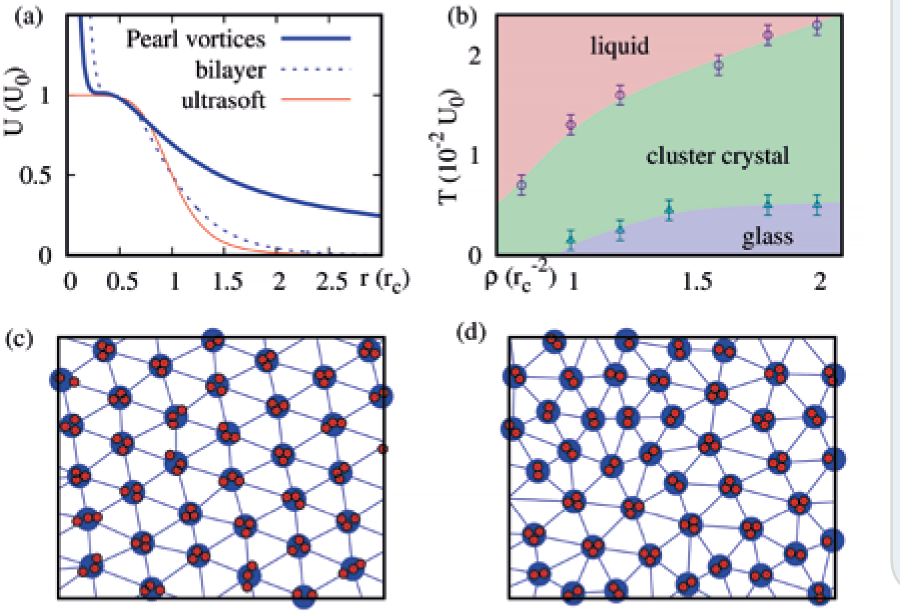

(a) Examples of types of "ultrasoft" interactions in a system of vortices: interactions for vortices in 1.5 superconducting films [thick blue line], interactions for vortices in 1.5 bilayer superconductors [dashed blue line], and a generic ultrasoft potential [red line].

(b) Dynamic phase diagram of a two-dimensional model of vortices in 1.5 superconducting films [see panel (a)], as a function of rescaled density ρ and temperature T. Circles and triangles indicate the liquid-to-crystal and the glass transition temperatures, respectively. (c) Snapshot of a crystal configuration after quenching a monodisperse vortex system with the potential for vortices in 1.5 superconducing films [blue line in panel (a)], for density $r^2_c\rho = 1.6$ at temperature $T = 1.8 \times 10^{-2}U_0$ [see panel (b)]. Single vortices (red circles) group into clusters (blue circles), blue lines join nearest neighboring clusters as obtained by Delauney triangulation. (d) Same as (c) for the glass phase at density $r^2_c\rho = 1.6$ and temperature $T = 0.2 \times 10^{-2}U_0$.

AWARDS

2017 prize « Espoirs de l'Université de Strasbourg »

This prize rewards the quality of the work and background of young researchers of the University of Strasbourg. This year, ten laureates, six men and four women were selected for the originality of their work and the dynamism in the implementation of their promising research.
Our colleague, **Jean-François DAYEN** (second from the right in the photo) is one of the laureates for his research on nanotechnology.

European Research Council (ERC) Consolidator Grants

Guillaume SCHULL is one of the 2017 CNRS laureates for his Project on Atomic-scale physics of single-photon sources. His project consists in using a new type of optical microscopy approach based on a scanning probe instrument, to probe the quantum properties of molecular chromophores with an atomic-scale precision.

Single electron devices based on graphene/nanoparticles heterostructures

The rise of graphene, followed by the more recent (re)discovery of the vast family of 2D transition metal dichalcogenides, has fueled an unprecedented inter-disciplinary research effort, at the interface between physics, chemistry and engineering.
A fast growing research area is now exploring the ability of graphene to promote the growth and self-assembly of nanoparticle on its surface. From the interaction of graphene with these nano-objects, new properties are expected to emerge. Such assemblies of nanoparticles, almost monodispersed in size, offer many possibilities in the field of nanosciences and in particular as regards catalysis, optoelectronics, data storage, and quantum technology.

From the electrical point of view, at the ultimate scale of the nanoparticle, the energy quantization of the electric charge induced by the Coulomb blockade (a phenomenon related to the electrostatic repulsion generated by the charges present on the nano-object), causes the appearance of clearly marked oscillations in the voltage response of the cluster conductance. These well-known effects are at the origin of single-electron transistors, which are considered as a serious alternative to the CMOS transistor technology and potential building blocks for *quantum electronics* and quantum information processing. However, the technical difficulty of manufacturing and then connecting a single cluster of a few nanometers in diameter, and the fact that on a larger scale, i.e. when several clusters are connected, the special properties of Coulomb blockade transport are blurred by averaging across the different clusters, represents important technological locks.

IPCMS researchers have recently discovered a growth process allowing the realization of assemblies of aluminum based nanoparticles, self-organized on graphene, buried within a nanometer thick alumina insulating layer. The low-temperature study of the resistance of this material revealed the existence of remarkably robust Coulomb oscillations, even for component sizes of 100 μm² incorporating millions of nanoparticles (Fig 1c,d). Surprisingly, the devices behave as if a single cluster were causing the measured transport. The reproducibility and the robustness of this transport have been confirmed by the researchers in several architectures of components and for different sources of graphene (graphene CVD on nickel, exfoliated).

These results open up new opportunities for the integration of graphene hybrid materials into nanoelectronics and spintronics devices, and are leading the way towards new architectures of single electron transistors and quantum electronics devices. ■

F. Godel et al., Adv. Mater., **2017**, 29, 1604837.
DOI: 10.1002/adma.201604837

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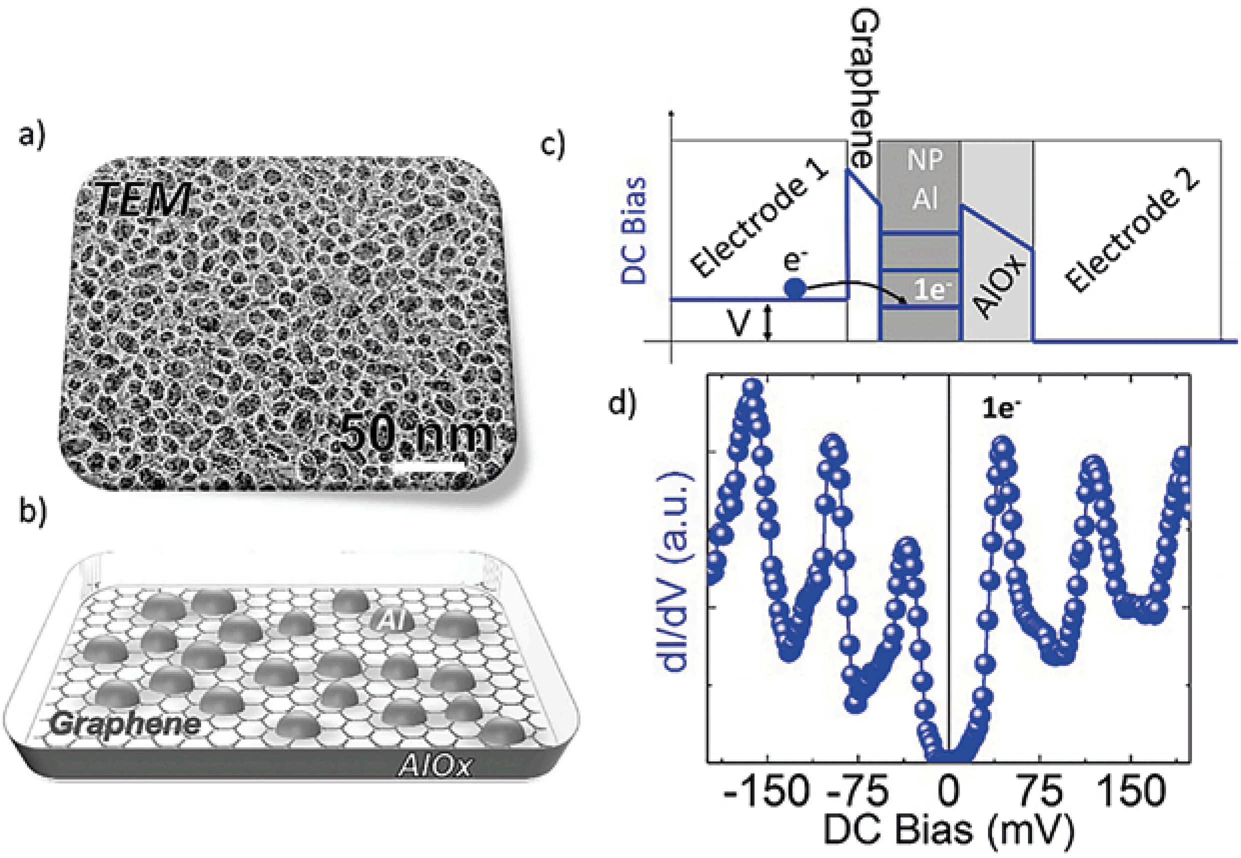

Figure :
a) TEM image of the graphene-nanoparticle hybrid. Aluminum based nanoparticles with average diameter of 6 nm appear clearly. (b) 3D schematic view of the graphene-nanoparticles hybrid. (c) Diagram of electronic transport through the graphene/Al/AlOx/Metal stack based on the energy levels (blue lines) with respect to the different layers. (d) Conductance versus voltage curve measured at 1.5 K in a 1 μm² large junction with hundreds of nanoparticles contacted in parallel. Robust and well defined conductance oscillations are observed.

Coherence- and spin-dynamics in high quality gallium nitride quantum well

We have investigated the dephasing time of free excitons in a high-quality single GaN/AlGaN quantum well by means of spectrally resolved and time-resolved four-wave mixing (FWM) as a function of temperature and excitation-power density. As a matter of fact, despite their technological importance for optoelectronics and high-power high-frequency electronics, such data concerning III-nitride quantum heterostructures were lacking, even though such information would prove extremely useful to understand the spin physics of excitons in these systems.

The measurement of the dephasing time T_2 allows one to determine collision rate $(\tau_C)^{-1}$ of excitons with phonons, within the exciton population, and with crystal defects. It provides valuable information about the mechanisms that govern spin relaxation, where spin precession is expected between two interaction events. We measured a spin-relaxation rate $(\tau_S)^{-1}$ that remains constant for temperatures <80K and is independent of the T_2 time. From a comparison with the dephasing rate $(T_2)^{-1}$, we conclude that spin relaxation is affected by motional narrowing. We can even estimate the electron-hole exchange interaction that governs the exciton-spin precession to 0.45 meV at T = 10 K, with the frequency of the latter increasing with temperature.

Our results use both optical coherence decay measurements, through FWM experiments, and spin-relaxation measurements, by polarized pump-and-probe experiments. This combination appears as a very powerful in order to reveal the microscopic mechanisms that govern the spin dynamics. The ability to perform such measurements in GaN-based heterostructures at short wavelengths in this family of technologically mature semiconductors opens promising perspectives in view of fabricating advanced nanophotonic devices based on these compounds. As an illustration, precise knowledge of the dephasing properties of nanoemitters would certainly prove extremely valuable, e.g., when embedding quantum dots in nanocavities exploiting cavity quantum electrodynamics phenomena such as the enhancement of the spontaneous emission rate via the Purcell effect. ■

M. Gallart, M. Ziegler, O. Crégut, E. Feltin, J.-F. Carlin, R. Butté, N. Grandjean, B. Hönerlage, and P. Gilliot,
Physical Review B, **2017**, 96, 041303(R).
DOI: 10.1103/PhysRevB.96.041303

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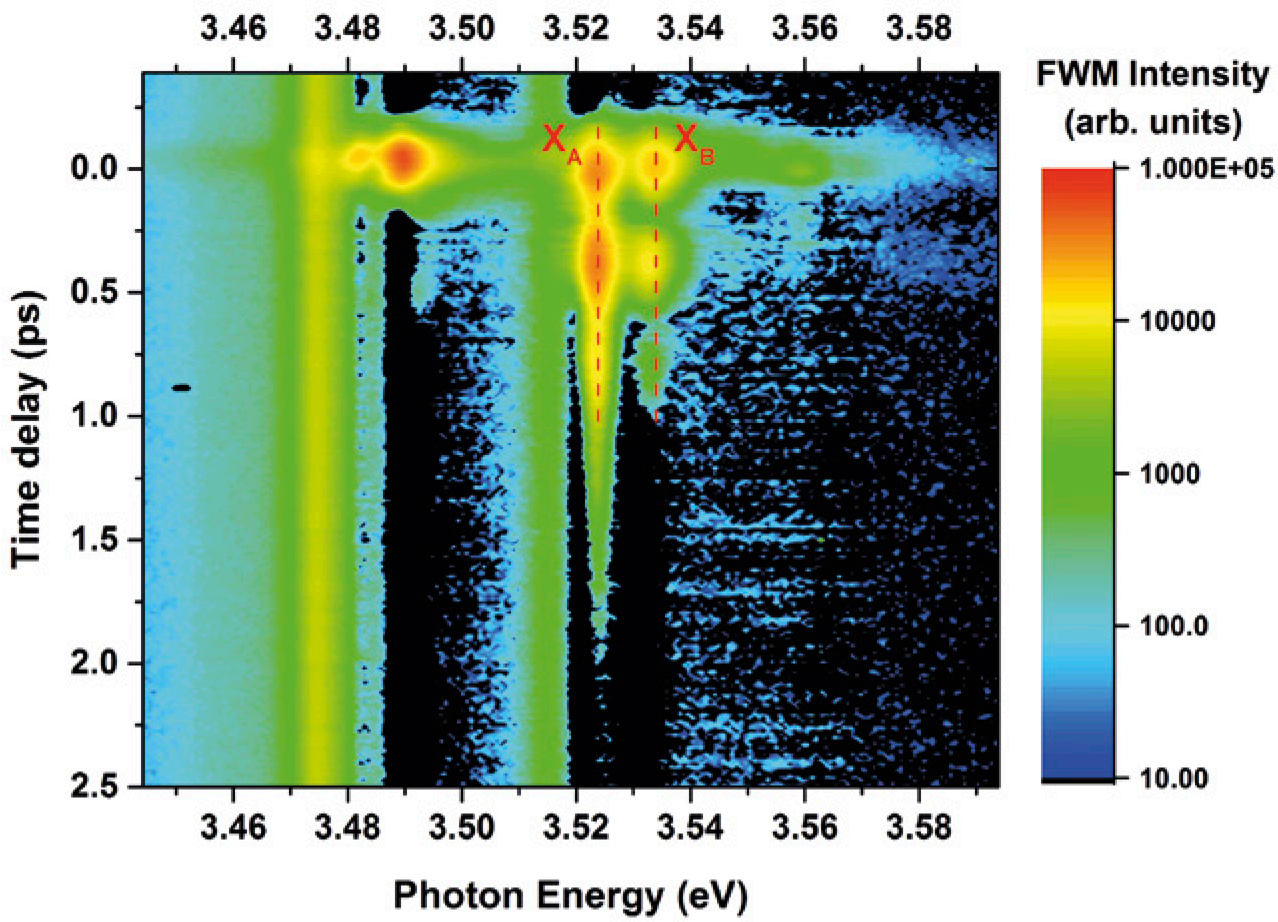


Intensity of the time-resolved and spectrally resolved FWM signal of a GaN/$Al_{0.05}Ga_{0.95}N$ quantum well at low temperature (T = 7K). The vertical scale corresponds to the time delay between the pump-and-probe pulses while the horizontal scale displays the photon energy. X_A and X_B indicate the two resonances related to the non-degenerate QW excitons (heavy- and light-hole excitons resonances, respectively).

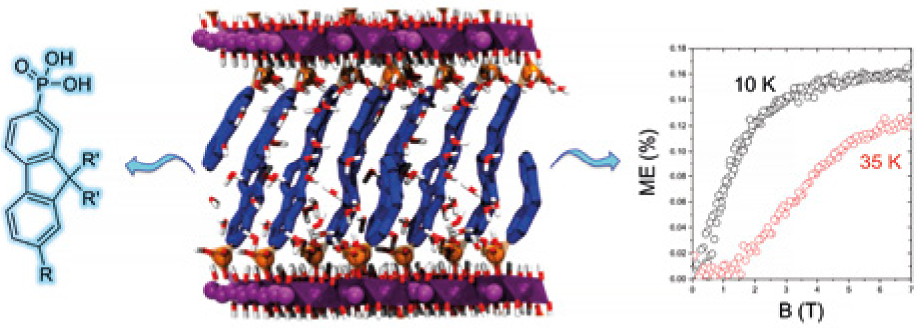
◆ Layered Simple Hydroxides functionalized by fluorene-phosphonic acids : synthesis, interface theoretical insights and magneto electric effect

The field of multiferroic and magneto electric materials is currently mainly covered by oxides, but it appears that the still scarcely explored chemical hybrid approach may present various advantages with respect to the solid state chemistry approach, in terms of synthesis and possibility to modulate rather easily the structures and properties.

Within the frame of an ANR program (HYbrid Multiferroic Nano-materials, contract N° ANR-14-CE07-0004-01, collaboration between IPCMS (Strasbourg), CRISMAT (Caen) and CEMCA (Brest)), we have investigated the functionalization of layered magnetic hydroxide hosts by phosphonic acids, in order to combine magnetic and dielectric properties within a single hybrid material.

Copper- and cobalt-based layered simple hydroxides (LSH) have been successfully functionalized by a series of fluorene mono- and di-phosphonic acids, using anionic exchange reactions and a preintercalation strategy. The lateral functionalization of the fluorene moieties has only little impact on the overall structure of the obtained layered hybrid materials but it influences the organization of the molecules within the interlamellar spacing. For bulky fluorene (9,9-dioctyl derivative), luminescence is preserved when inserted into copper and cobalt hydroxides, whereas it is completely quenched for the other fluorenes, which indicates different packings of the molecules within the interlamellar space. Detailed characterization of the internal structure and chemical bonding properties for copper- and cobalt-based hybrids is performed *via* ancillary experimental techniques. For the copper-based LSH class, for which more elusive findings are found, first-principles molecular dynamics simulations unravel the fundamental stabilizing role of the H-bonding network promoted within the local environments of the fluorene mono- and di-phosphonic acids.

The cobalt series of compounds constitute a new class of hybrid magnets, with ordering temperatures ranging from 11.8 K to 17.8 K. In addition, it shows a very clear magnetoelectric effect (magneto-capacitance). This effect appears above a threshold magnetic field which is null below the magnetic ordering temperature, and it persists in the paramagnetic regime till about 110 K.



These finding pave a novel way to fill the critical need for a deeper understanding of the influence of the organic and inorganic subcomponent interactions for innovative magnetoelectric hybrid compounds with potential ferroelectric properties. ■

Q. Evrard, Z. Chaker, M. Roger, C. M. Sevrain, E. Delahaye, M. Gallart, P. Gilliot, C. Leuvrey, J.-M. Rueff, P. Rabu, C. Massobrio, M. Boero, A. Pautrat, P.-A. Jaffrès, G. Ori, G. Rogez,
Adv. Funct. Mater., **2017**, 27, 41, 1703576.
DOI: 10.1002/adfm.201703576

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IPCMS News

◆ Fluorescence microscopy with sub-molecular resolution

Addressing the optical properties of single-atomic or molecular sources has become a major research topic that can be traced back to the first observation of a single-molecule in an absorption spectrum by Moerner and Kador in 1989, rapidly followed by the first measure of the emission of a single-molecule by Orrit and Bernard in 1990. These observations have triggered the development of new methods aiming at increasing the resolution in fluorescence microscopy experiments with major consequences in chemistry, biology and medicine. Eric Betzig, Stefan W. Hell and William E. Moerner receive the Nobel Prize in Chemistry in 2014 for the development of these “super-resolved fluorescence microscopy" approaches. However, even these very powerful techniques are limited in resolution at the very best to approximately 10 nm. This is the range where charge and energy transfers between a single-molecule source and its environment occur.

In our recent article we developed an original approach that allows us probing and gaining control on a single-molecule emitter with sub-nanometric precision. We used the tunneling current traversing the junction of a scanning tunneling microscope (STM), working under vacuum and at cryogenic temperatures, to excite the fluorescence spectra of individual Phthalocyanine molecules deposited on salt-covered silver surface. The recorded emission spectra reveal a sharp zero phonon line as well as several vibronic peaks that can be considered as a reliable fingerprint of the molecule. We could then record how each of these vibronic features varies as a function of the position of the STM tip with respect to the molecule, providing vibronically resolved fluorescence maps with sub-molecular resolution. These results question our understanding of what is the fluorescence spectrum of a molecule and open the way to measurements where optical microscopy reach atomic-scale resolution. ■

B. Doppagne, M. C. Chong, E. Lorchat, S. Berciaud, M. Romeo, H. Bulou, A. Boeglin, F. Scheurer and G. Schull, Physical Review Letters, **2017**, 118, 127401.
DOI: 10.1103/PhysRevLett.118.127401

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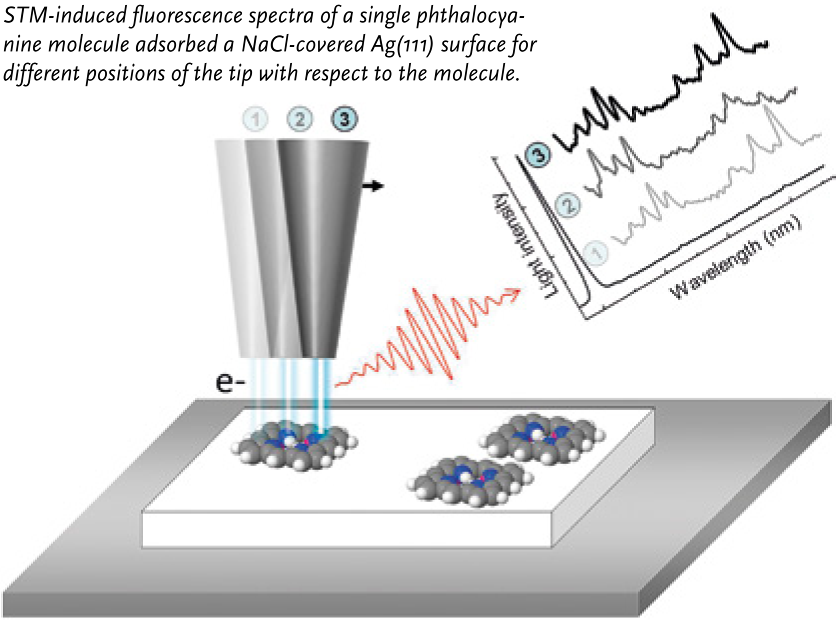
STM-induced fluorescence spectra of a single phthalocyanine molecule adsorbed a NaCl-covered Ag(111) surface for different positions of the tip with respect to the molecule.



NANOTRANSMED – Innovations in nanomedicine: from diagnosis to implants

NANOTRANSMED (2016-2019) is a cooperation project implemented by 8 laboratories from the Upper Rhine Region, among them IPCMS, for a total budget of 4 590 853,50 € including 1 978 559 € from ERDF and 633 735,50 € from the 5 Swiss partners. NANOTRANSMED enables French, German and Swiss scientists to cooperate on improving patients' care by early diagnosis of diseases, personalized treatment and solutions to fight against nosocomial (healthcare-associated) infections.

The scientists of the NANOTRANSMED consortium rely on a "multifunctional" approach, based on the cooperative dendritic materials technology. The aim is to graft polyvalent branched molecules – dendrimers – on a substrate in order to create multi-functional nano-objects or surfaces. These dendritic molecules can be synthetized in a controlled and iterative manner. Moreover, their architecture can easily be tuned to solve different issues as regards to biocompatibility, bioelimination or targeting.

The researchers aim at designing, developing and mastering biocompatible and innovative nano-objects (circulating probes or implant coatings) by conferring them i) specific dual-targeting capacities, ii) antibacterial properties, iii) enhanced in vivo stability, iv) furtivity and v) reduced toxicity.

The objectives of the project in terms of improved patients' care thanks to nanomedicine are the following:

- Establish an early, fast and reliable diagnosis by improving the targeting efficiency of imaging probes to diagnose diseases such as cancer or inflammation,
- Deliver a personalized treatment, by the development of theranostic nano-objects, i. e. capable of combining efficiently targeted treatment and therapy monitored by imaging,
- To fight nosocomial infections linked to invasive interventions requiring a medical device (catheter, probes, vascular or cardiac implants ...). Many implants must display smart antibacterial surfaces to avoid microbial colonization.

These new probes and implant coatings will be directly used at the end of the project by the Hospitals of Basel and Freiburg associated to this project and conducting preclinical studies, as well as by any company, biotech or equipment supplier in the field of diagnosis and therapy or implantology (valves, pacemakers, cardiac defibrillators).

Two symposia will be organized in the framework of the project at the E-MRS Spring Meeting in June 2018 in Strasbourg: Symposium E "Nanotechnology for targeted personalized medicines and theranostics" and Symposium F "Advanced biomaterials: elaboration, nanostructure, interfaces with tissues".

NANOTRANSMED is an INTERREG V Upper Rhine project co-financed by the European Regional Development Fund.

www.nanotransmed.eu

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◆ *Magnetic permeability control in dendronized Manganese-Zinc-Ferrite nanoparticles*

Magnetic NPs are attracting interest due to their potential applications in data storage, AC electromagnetic devices... Collective magnetic properties of NPs depend on their size, shape, chemical composition, as well as their assembly and interparticle distance (dipole–dipole interactions). Strong dipolar interactions alter the energy barriers for the thermal relaxation of NP magnetic moments, thus the ferromagnetic resonance (FMR) frequencies. FMR is a phenomenon in which the external magnetic field energy is absorbed by magnetic materials when the spin precession frequency of the magnetic domains coincides with the external AC magnetic field frequency. FMR thus limit the operable frequency range of magnetic materials in AC magnetic devices, and increasing their range to higher frequencies (i.e. radio frequency) is of great importance especially for the miniaturization of such devices. FMR frequencies can be tuned by manipulating the NPs interactions, and one way to overcome this limit is to engineer bulkiness within the ligand shell that will create the necessary steric bias. Dendritic ligands seem appropriate as they can finely tune bulkiness and engineer controlled interparticle distances.

Soft ferrite such as manganese zinc ferrite (MZF) NPs were chosen as a model system. A controlled increase in the interparticle distance was achieved after exchanging the oleic acid (**OA**) surface capping ligands with the phosphonic acid-terminated dendrons from 2.6 to ca. 4.5 nm (**G1**) and 5.0 nm (**G2**), as seen by TEM. The blocking temperature (T_B) of the NPs, where the ferromagnetic to superparamagnetic transition occurs, is significantly decreased from 111 K (**OA**) to 75 K (**G1**) and to 63 K (**G2**). Due to the increased interparticle distance, the dipolar interactions between NPs as well as the energy barrier for the thermally induced spin re-orientation decrease, hence the lower T_B. The decrease of the real part of the susceptibility (χ') from 9.5 (**OA**) to 4.5 (**G1**) and 2.4 (**G2**) is attributed to the reduction of the NPs volume fraction in the dry powder sample. Therefore, the magnetic field flux density in the sample becomes lower after ligand exchange. The superparamagnetic–ferromagnetic relaxation frequency corresponds to the frequency at which χ'' value is maximum. The χ'' value of MZF@OA reached a maximum at 234 MHz. After ligand exchange, χ'' reached a maximum value at 342 MHz for (**G1**), and a slight increase without a maximum point up to 500 MHz (**G2**), suggesting that the maximum is higher than 500 MHz. This frequency increase results from the reduced dipolar interactions following ligand exchange, and indicates that the operable frequency range is extended toward higher frequencies. The significant reduction in χ'' gives rise to a huge change in the energy efficiency of the material. These results clearly demonstrate the effect of the inter-particle spacing on the AC magnetic properties of MZF NPs at radio frequencies and support the suitability of our approach for employing NPs in AC magnetic devices. Further optimization of ligand exchanged NPs will be needed to achieve a higher real part of permeability for magnetic applications.

Light-mediated straintronics can therefore be an alternative way for multi-functionality between spins, electrons and electric charges where the photostriction was a missing property (see figure). ■

D. Jishkariani, J. D. Lee, H. Yun, T. Paik, J. M. Kikkawa, C. R. Kagan, B. Donnio and C. B. Murray,
Nanoscale, **2017**, 9, 13922-13928.
DOI: 10.1039/c7nr05769e

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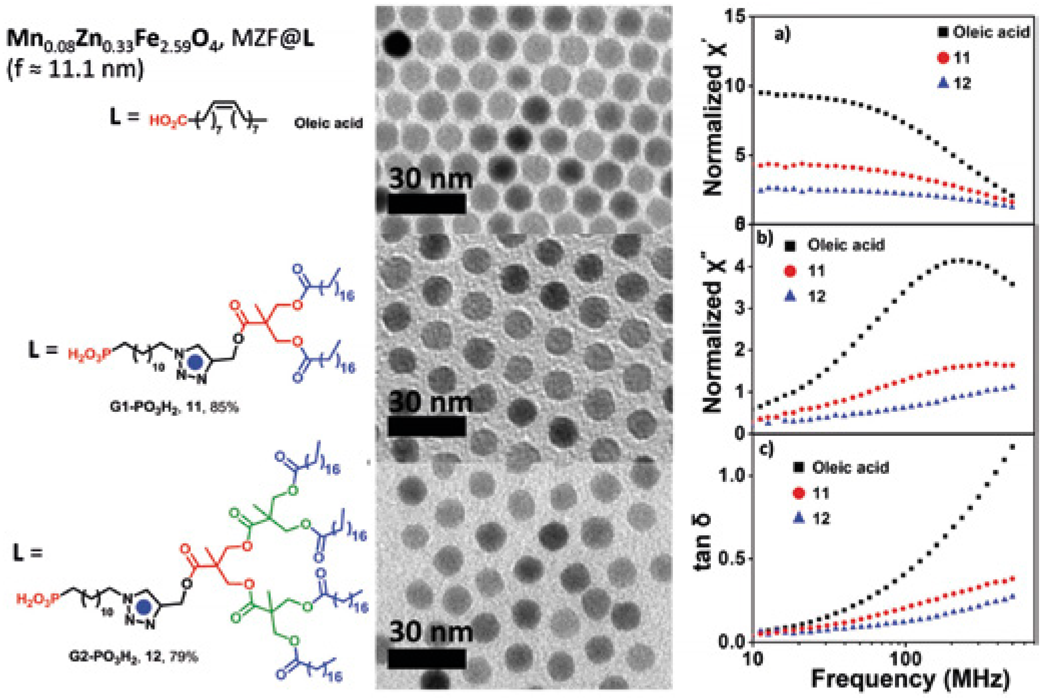

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