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Editorial

This issue of *IPCMS News* highlights some recent achievements of the institute in the fields of spintronics, magneto-optics, biophysics and materials for health. It shows new results on nanocontacts including single molecules (oriented C₆₀) to minimize the interface effects in spintronics devices and control the tunnel transport regime, the change of the electronic properties of molecules at the interface with a metal, the development of well oriented GaFeO thin films with a room temperature magnetic moment and a strong magneto-electric coupling. Further outcomes are on the magneto-optical control of molecules which are good candidates for memory devices, highly relaxing contrast agents for MRI based on iron oxide nanoparticles stabilized by dendritic molecules, and the quenching mechanism of tryptophans that are commonly used intrinsic fluorescence sensors for e.g. protein solvation dynamics or intra-protein charge transfer.

"Sino-French Graduate School for Photonics and Laser Science" : Strasbourg University, and in particular IPCMS, is one of four French partners in a newly created Sino-French network for academic exchange and research training in Photonics and Laser Science. With the agreement signed on nov, 3rd 2010, by the Wuhan National Laboratory of Optoelectronics at the Huazhong University of Science & Technology a Sino-French graduate school for joint PhD programs is created involving the French Universities Paris-11, Lyon 1, Angers and Strasbourg.



HUST vice president Prof. Q. Luo and S. Haacke (IPCMS) hand-shake after signature of the joint agreement. In the back, from left to right: R. Barillé (U Angers), F. Vallée and N. Del Fatti (Lyon), E. Cassan (Paris-11), T. Heiser and J.-Y. Bigot (Strasbourg), M. Lavroff and J. Fleck (French consulate Wuhan) and K. Yang (WNLO).

Marc Drillon
Director

<http://www.consulfrance-wuhan.org/Temps-fort-a-Wuhan-pour-l-activite-optoelectronique-et-les-technologies-lasers.html>

◆ The interface between a molecule and a ferromagnetic surface: a new eldorado for spintronics

Within a solid-state magnetic tunnel junction, electron tunnelling from one ferromagnetic electrode to the other results from two considerations. First, the hybridization at the interfaces of certain wavefunctions allows the electron to transition from a diffusive transport regime in the metal toward the tunnelling regime. Second, the tunnelling transmission of these wavefunctions across the ultrathin insulator is determined by the electronic structure of the latter.

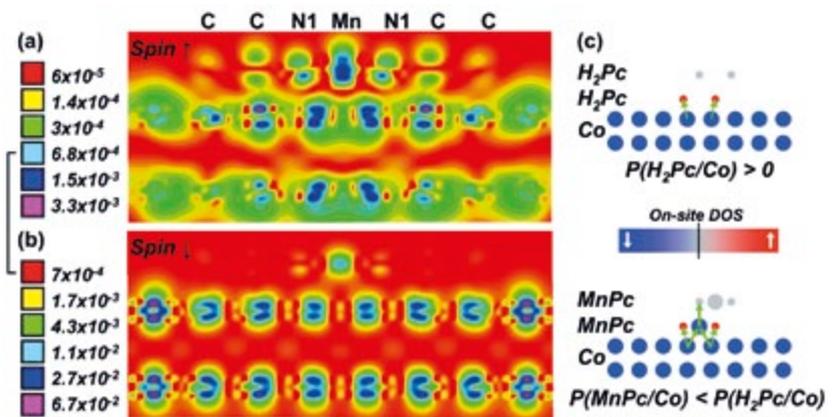
A close collaboration between several IPCMS teams and the Wulfhekel group from the KIT (Karlsruhe) has recently evidenced novel spintronic properties at the interface between a manganese phthalocyanine molecule and a Co(001) surface. Indeed, synchrotron radiation experiments, performed using the TBT equipment operated by the IPCMS at the Swiss Light Source, have revealed an important modification of the molecule's electronic structure: due to a strong hybridization with Co, the first molecular plane acquires a metallic character. As a result, the effective tunnelling transport interface now appears between the first and second molecular planes.

Ab-initio calculations show that, due to charge transfer resulting from the molecule-Co bonds, the spin polarization P of this effective interface is quite different from that of a bare Co surface with a negative P (ie with spin \downarrow dominated transport). While P remains negative on the central Mn site of the molecule, P changes sign to become positive on the C and N molecular sites (ie with spin \uparrow dominated transport). Since wavefunctions with Mn character may favorably transmit across a MnPc barrier, it should therefore be possible to control the amplitude and sign of the tunnel-

ling spin polarization simply by omitting or not the central molecular site. This ease of spintronics control is unparalleled in inorganic spintronics. ■

Impact on interface spin polarization of molecular bonding to metallic surfaces, S. Javaid, M. Bowen, S. Boukari, L. Joly, J.-B. Beaufrand, X. Chen, Y.J. Dappe, F. Scheurer, J.P. Kappler, J. Arabski, W. Wulfhekel, M. Alouani, E. Beaurepaire, Phys. Rev. Lett. **105**, 077201 (2010)

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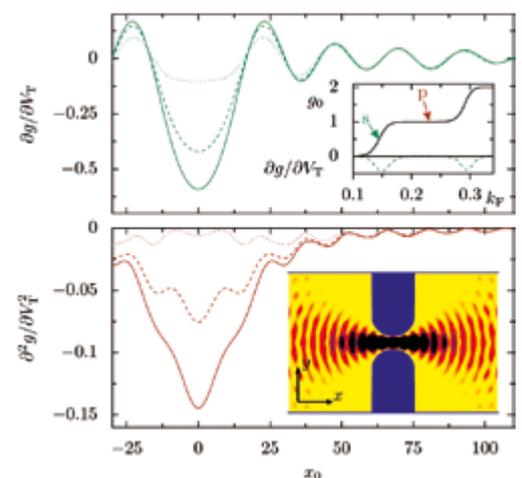
◆ What Is Measured in the Scanning Gate Microscopy of a Quantum Point Contact?

Peeping electrons going by is one of the most fascinating issues of Quantum Mechanics. Attempting such an experiment in a solid-state environment, like in a two-dimensional electron gas (2DEG), involves subtle fundamental ingredients such as decoherence and many-body effects, as well as cross-talks and other technical limitations. The Scanning Gate Microscopy (SGM), developed in recent years, is an attempt to measure the electron flow. This technique is based on the measurement of the conductance through the sample. A local gate electrode, typically a charged tip of a scanning tunnelling microscope, creates a local perturbation in the 2DEG, and leads to a change of the measured conductance that depends on the position of the tip. These conductance changes have often been interpreted as a mapping of the electron current density along a nanostructured device. In order to understand what is really meas-

► *Top: The linear coefficient of the conductance change with the tip voltage as a function of the distance of the tip from a QPC that is tuned to the first conductance step (point s of the conductance curve of the inset).*

► *Bottom: The second order coefficient when the QPC is tuned to the first plateau (p). Inset: Grayscale plot of the conductance changes as a function of tip position with respect to the QPC.*

ured by such conductance changes, we developed a systematic perturbation theory for the effects of a local perturbation on the conductance of a nanostructured 2DEG. In agreement with the dictates of Quantum Mechanics, our analysis shows that the SGM measurements in a phase-coherent nanostructure are not given by a local quantity. The resulting expres-



sions for the conductance change in first and second order in the perturbation contain matrix elements of the perturbing potential with two scattering states impinging from opposite electrodes. The case of a Quantum Point Contact (QPC) is of particular interest because a number of experimental SGM data is already available. A QPC is a narrow con-

striction in the 2DEG that can lead to conductance quantization in units of $2e^2/h$. When one varies the Fermi wavelength, the conductance of the QPC exhibits steps and plateaus (see the inset in the upper part of the figure). We find that the first-order contribution for weak tip voltages is significant only on the conductance steps (upper figure; dashed line of the inset). Therefore, the

second-order correction (lower figure) is the dominant one on the plateaus. The latter contribution is always negative, exhibits fringes, and has a spatial decay consistent with SGM experiments on QPC. ■

This work has been performed in collaboration with Steven Tomsovic (Washington State University, Pullman, USA).

What is measured in the scanning gate microscopy of a quantum point contact?, R.A. Jalabert, W.Szenc, S. Tomsovic, D. Weinmann, Phys. Rev. Lett. 105, 166802 (2010)

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IPCMS News

◆ Dendronized iron oxide nanoparticles as contrast agents for MRI

Some of the most significant and promising applications for inorganic nanoparticles (NPs) lie in the fields of biology and biomedicine. Still, control of the size and stability of biofunctionalized particles in biological media are key issues in the development of inorganic nanoparticles for such applications. Due to their magnetic properties and in particular their very high transverse relaxivity, superparamagnetic iron oxide nanoparticles (SPIO) with appropriate surface chemistry can be used in numerous *in vivo* applications such as MRI contrast enhancement, hyperthermia treatment, cell sorting, drug delivery, immunoassay and tissue repair. In all of these fields, it is necessary (mandatory) to engineer the surface of SPIO NPs not only to improve biocompatibility, solubility and stability in physiological media but also to ensure a narrow particle size distribution after decoration and preserve good magnetic properties. In addition, small-sized particles (below 100 nm) have a good bio-distribution after intravenous injection.

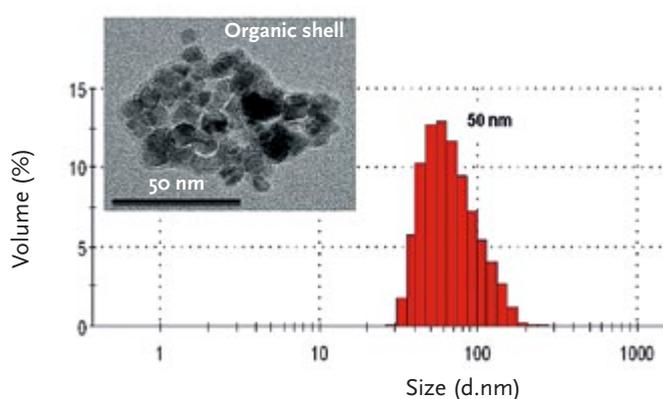
With the double objective of increasing the grafting efficiency and providing the possibility of tailoring the characteristics (morphology, functionalities, physico-chemical properties...) of the organic coating, we proposed a strategy based on a combination of dendritic and phosphonate approaches. (i) The choice of dendritic molecules is motivated by the fact that they are discrete entities leading to thinner coatings than polymers. Moreover, they display relevant characteristics (size, hydrophilicity, molecular weight...) which can easily be tuned as a function of their generation. Additionally, they allow different and reproducible poly-functionalization at their periphery. (ii) The choice of phosphonic acid as a coupling agent is justified by previous studies which have evidenced that it allows a significantly higher grafting rate and a stronger binding than a carboxylate anchor.

This strategy has been validated by comparing the highly relaxing colloidal water suspensions so elaborated to polymer-dec-

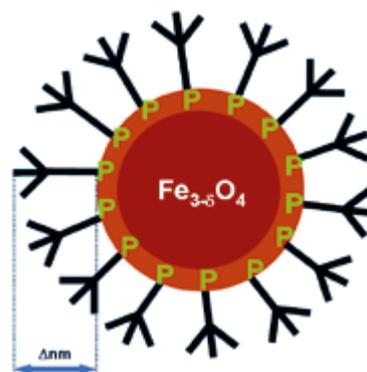
orated commercial SPIO-based contrast agents. From T_1 and T_2 values measured by relaxometry at 1.5 T (room temperature) as a function of concentration, relaxivity values as high as $349 \text{ mM}^{-1} \text{ s}^{-1}$ for r_2 and $10 \text{ mM}^{-1} \text{ s}^{-1}$ for r_1 , with a 44.8 R_2/R_1 ratio, were obtained for our nanoparticles with the smallest hydrodynamic diameter. These values are significantly higher (1.5 times superior) than those of commercial polymer-decorated nanoparticles.

In vitro relaxivity measurements at 7 T and 37°C were performed on one microliter samples containing increasing concentrations of dendronized materials dispersed in water. Enhancement contrast ratios (EHC) were calculated for our dendronized NPs in reference to water $[\text{EHC}(\%)_w]$ and to EndoremTM at equivalent iron concentration $[\text{EHC}_{\text{EN}}]$. The very high EHC values obtained for our NPs confirmed their very high contrast power even at high magnetic field (7 T). For instance, on MR T_{2w} image at 7 T, EHC values are from 15% to 75% higher than those obtained for EndoremTM.

All these analyses confirm the great interest in grafting small dendritic molecules through a phosphonate anchor in order to stabilize iron oxide suspensions by electrostatic and steric repulsions. Such hybrid and biocompatible nano-objects open a new route for the development of highly



◀ TEM micrograph (inset) of the decorated NPs and particle size distribution in volume of NP water suspension at physiological pH.



▲ Schematic representation of dendronized iron oxide nanoparticles through phosphonate anchor.

relaxing contrast agents displaying a quite satisfactory R_2/R_1 ratio even at high field. In addition, we expect that further grafting of biological effectors through the dendritic platform will allow efficient targeted imaging and therapy.

Dendronized iron oxide nanoparticles as contrast agents for MRI, B. Basly, D. Felder-Flesch, P. Perriat, C. Billotey, J. Taleb, G. Pourroy, S. Begin-Colin, Chemical Communications, 2010, 46, 985-987.

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◆ Welcome to the Nanotrench !

New breakthroughs envisioned for molecular and nano electronics in the area of information storage and processing have motivated a significant boost to both fundamental and applied research in this area. Since first pioneer work, the field is entering now in a mature stage. It has become increasingly clear that the realization of robust single molecule or single nanoparticle devices exhibiting reliable properties is a very challenging experimental and technological task. Devices based on multiple connected nanoparticles or multi-scale approach, should be less sensitive to such artifacts. The larger number of interconnects ensures device robustness through ensemble averaging. Moreover, nanomaterials commonly exhibit high sheet resistance values, with additional exponential increase under cooling, due to hopping transport or Coulomb blockade associated transport.

There is therefore a need for developing new configurations of electrode interconnects with high aspect ratio (HAR), exhib-

iting good interface resistance, and processed with standard industrial tools. We have recently demonstrated the easy fabrication of a new type of electrodes geometry, of relevance for nanoelectronic devices, that we refer to as "nanotrench". The process uses standard optical lithography techniques and allows parallel processing of many HAR junctions, making it scalable and industrially relevant. Aspect ratio up to 10^4 is demonstrated (Fig.a), with electrode of $100\mu\text{m}$ width, and inter-electrode gap down to 20 nm (Fig.b). Current leakage below 1 pA was systematically observed.

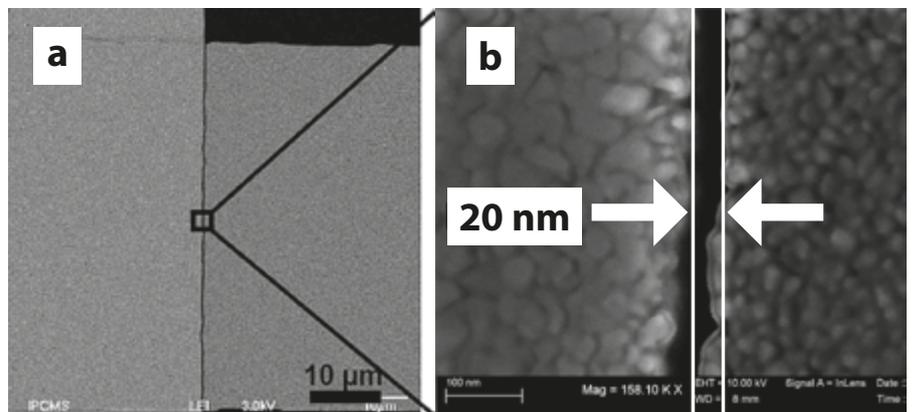
We demonstrated the suitability of these nanotrenches as electrical interconnects for addressing micro and nano particles by realizing several circuits with integrated species. Trapping of conducting microspheres with low impedance contact between the spheres and the electrodes on each side of the nanotrench were successfully obtained (Fig.c), demonstrating good quality electrical interconnects, even though a wet process was used (V. Faramarzi, PhD). Highly resistive magnet-

ite-based nanoparticles networks deposited by Longmuir-Blodgett technique (Fig.d) demonstrated the advantage of the high aspect ratio of the nanotrenches for providing access to electrical properties of highly resistive materials down to low temperature (collaboration with DCMI). Work is under progress on these systems, where magnetoresistance up to 45% has been recently measured on Fe_3O_4 nanoparticles contacted into nanotrench, demonstrating the interest of this geometry for nanoparticles spintronic based device. ■

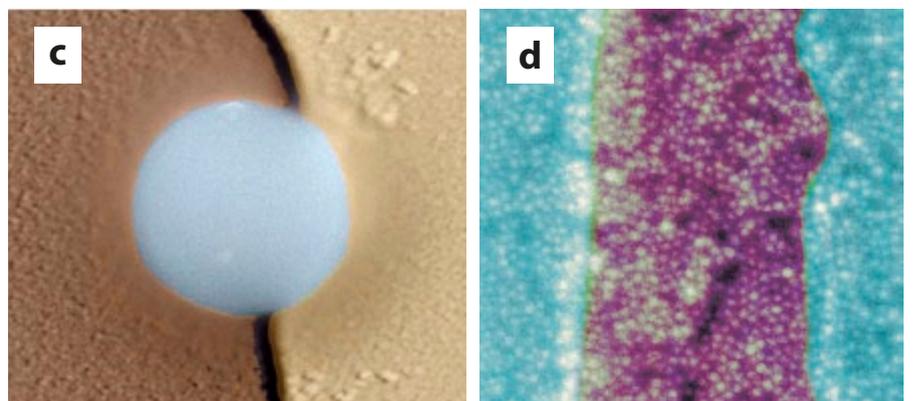
Nanotrench for nano and micro particle electrical interconnects, J.-F. Dayen, V. Faramarzi, M. Pauly, N.T. Kemp, M. Barbero, B.P. Pichon, H.Majjad, S. Begin-Colin, B. Doudin, Nanotechnology, 21, 335303 (2010).

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a) Nanotrench, (b) detail of a 20 nm gap. ▶



(c) Half gold coated microsphere trapped over the nanotrench electrodes, (d) Fe_3O_4 nanoparticles monolayer deposited into the nanotrench. ▶



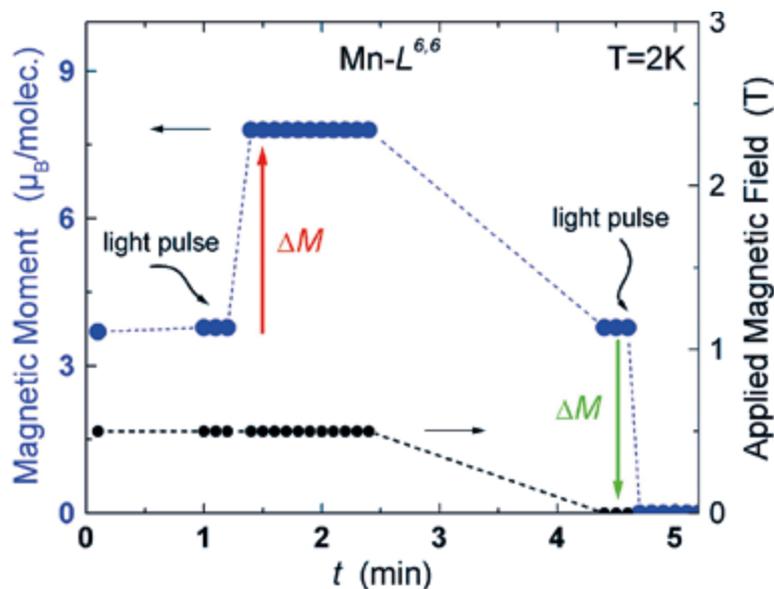
◆ Magneto-optical control of a Mn12 nano-magnet

Research in molecular electronics aims at using molecules as a replacement for silicon components. Firstly, because of the huge gain in density that would result. Secondly, because molecules are multifunctional, enabling the simultaneous presence of several properties on a single nano-object. Single-Molecule Magnets (SMM) are paramagnetic molecules with a non-zero and usually large individual spin S . Under appropriate field and temperature conditions a stable magnetization in the $+S$ or $-S$ state can be retained for long periods of time (years). Such molecules which behave as nanomagnets maybe viewed as the ultimate storage element in a magnetic memory. Ongoing research tries relying on the quantum nature for the fabrication of the qbits which would form the heart of a molecular computer. In both cases there is an obvious need for a means of controlling the magnetic or quantum state of the SMM.

In collaboration with E. Rivière (ICMMO, Orsay, France), we recently discovered that irradiation at optical wavelengths can actually trigger at will an increase or a decrease of the magnetic moment of a SMM, even though the energy of the photons does not correspond to a precise electronic or molecular spin transition. Using the proper light and field sequence, the magnetization of the system can be driven between at least three different stable states. The consequences of these observations are manifold: they bring a means of controlling molecular magnets, open prospects in the field of quantum computing, and may enable the realization of coherent microwave sources through stimulated superradiance.

In a typical experiment, the demagnetized sample is prepared in a magnetization state $M(H)$ by applying a field H ($\mu_0 H < 1.8$ T). If, at this given non-zero applied field, a light pulse is sent the magnetization is seen to *increase*. This M increase depends on both H and the magnetic history of the sample, reaching quite large values when the sample comes from $-M_{\text{Sat}}$. We emphasize that since the sample is on its first magnetization curve it is impossible to get an increase of M with an increase of temperature, which proves that the observed effects are not thermal. When another light pulse is sent, the magnetization drops to zero. Symmetrically, if the magnetization is first saturated and H then ramped down, the action of the light pulse is to decrease M .

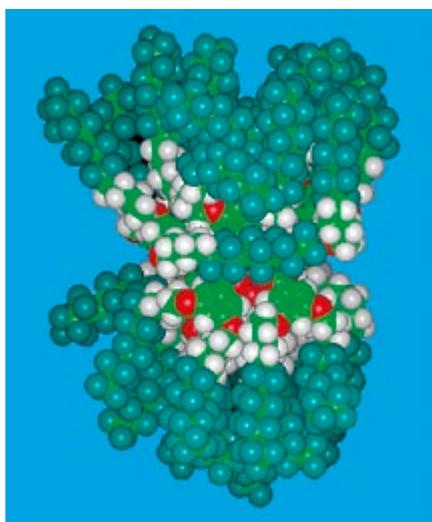
Performing pump-probe experiments or Raman scattering may yield valuable insights on this system. Given the fact that a single 5 ns light pulse can fully demagnetize the sample in its remnant state, it could be of interest to endeavour observation of stimulated emission of radiation (superradiance) in the microwave region. These results also open interesting perspective in the field of quantum computing, the addressing of qbits being today one of the major hurdles. ■



The observation that a short light pulse can very efficiently demagnetize a SMM is of primordial importance and worth understanding. Indeed it has strong implications for experimenters trying to probe the magnetic properties of SMM on surfaces using techniques such as XMCD, where high energy (~ 600 eV) photons are employed.

Magneto-optical control of a Mn12 nano-magnet, E. Rivière, B. Donnio, E. Voirin, G. Rogez, J.-P. Kappler, J.-L. Gallani, J. Mater. Chem., **20**, 7165- 7168, 2010.

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▲ Dendronized Single-Molecule Magnet

◆ Molecular contacts at hand

Investigations of the current flow passing through single atoms or molecules have revealed fascinating properties such as quantization of the conductance, reduction of the noise at a single atom contact, or the fabrication of suspended atomic chains between metallic electrodes. These studies have highlighted the major importance of the interfaces between the metallic electrodes and the contacted nano-objects. At the scale of a single molecule, the number of atoms at the interface and their positions can substantially modify the transport properties of the junction. So far, no experiment has provided control of these parameters.

In the present publication, Schull et al. use a method based on a scanning tunnelling microscopy to show that it is possible not only to determine but also to control the exact number of atoms at the interface between a single molecule (oriented C_{60}) and a metallic electrode (made of copper) while simultaneously monitoring the current passing through the junction. They discovered that the conductance of a molecule can vary by more than an order of magnitude depending on the cross-sectional area of the contact with the electrodes. By changing, one by one, the number of atoms in contact with the molecule, a crossover from a regime in which the conductance is limited by low injection of charges from the electrode to the molecule ("bad contact") to a regime in which the conductance is limited by the molecular states ("good

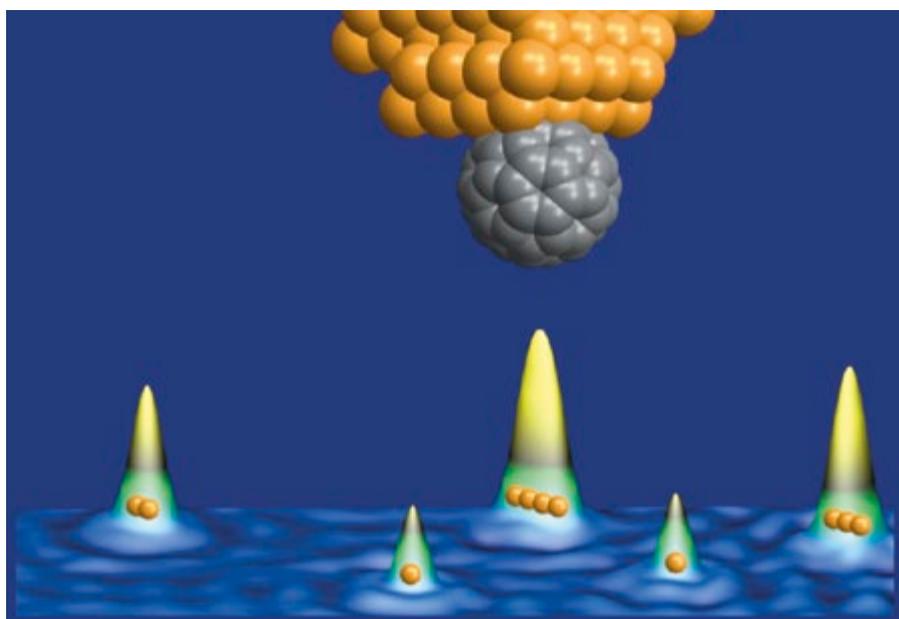
contact"). These results are surprisingly similar to what is observed in mesoscopic physics where the transport properties of promising conductors (carbon nanotubes, graphene nano-ribbons, carbon onions...) are intimately linked to the quality of the contacts to metallic electrodes. This suggests that single-molecule junctions can be used as model structures for the study of such materials. ■

Atomic-scale engineering of electrodes for single-molecule contacts, G. Schull, Th. Frederiksen, A. Arnau, D. Sanchez, R. Berndt, Nature Nanotech, Nature Nanotech. 6, 23–27 (2011)

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Graphical representation of a scanning tunnelling microscope tip with a C_{60} molecule fixed to its apex, above a copper sample with five atomic scale electrodes engineered atom per atom. ▼



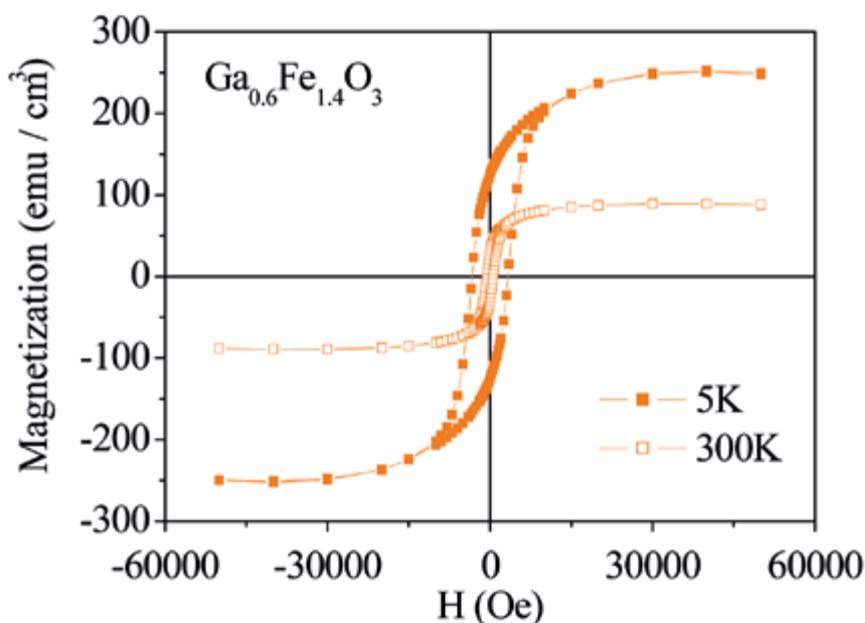
◆ Room temperature ferrimagnetic thin films of the magnetoelectric $Ga_{2-x}Fe_xO_3$

Magnetoelectric materials, thanks to the coupling existing between their magnetic and electric properties, allow the manipulation of magnetization by an electric field. Such materials currently receive considerable interest for they open new perspectives in terms of memory devices. Indeed, a magnetoelectric memory would combine the best of both the FeRAMs and MRAMs worlds. At the present time, the development of magnetoelectric memories appears to be limited by the scarcity of magnetoelectric materials and even more by the scarcity of magnetoelectric materials presenting a magnetic order at room temperature.

The magnetoelectric manipulation of magnetization has recently been proved experimentally at room temperature with a ferroelectric antiferromagnet, $BiFeO_3$ (BFO). This is the only material considered so far in the literature presenting both a magnetoelectric coupling and a magnetic order at room temperature. However, since the magnetic order is antiferromagnetic, the actual use of BFO requires the use of an extra layer to which it is magnetically coupled. This makes the fabrication of devices more complicated and the device itself subject to failures. It is therefore essential to make efforts in developing magnetoelectric materials presenting a non zero

magnetization at room temperature. With this aim in view, we have considered gallium ferrite $Ga_{2-x}Fe_xO_3$ ($0.8 \leq x \leq 1.4$) (GFO) with great interest. It indeed appears as the perfect alternative material to BFO in magnetoelectric memories : it is ferrimagnetic above room temperature for $x=1.4$ and pyroelectric with a strong magnetoelectric coupling.

While its bulk properties had been well established, GFO had been very little studied in thin films. Two important points had in particular to be addressed : (i) the possibility to obtain, as had been done in bulk, room temperature ferrimagnetic films by tuning the Ga/Fe ratio, and (ii) the possibil-



◀ M-H Hysteresis loops of a $Ga_{0.6}Fe_{1.4}O_3$ thin film at 5 K and at 300 K (in-plane measurements)

ity to deposit high crystalline quality GFO thin films on a conducting electrode. This last question conditions the actual use of GFO thin films in magnetoelectric devices. We have addressed both points and performed the elaboration of room temperature ferrimagnetic GFO thin films on conducting Pt(111) electrodes by pulsed laser deposition (PLD).

An optimization of the growth conditions of GFO thin films by PLD allowed us to obtain perfectly *b*-axis oriented $Ga_{2-x}Fe_xO_3$ films on yttrium-stabilized zirconia (001) (YSZ) substrates, for all *x* values between 0.8 and 1.4. The films present a strong anisotropy, the out-of-plane direction (GFO *b* axis) being a hard magnetization direction. The Curie temperature increases with *x*, as for the bulk, and a value superior to room temperature was obtained for $x=1.4$ ($T_C = 370$ K). The in-plane coercive field reaches the relatively high value of about 3200 Oe at 5 K for $x=1.4$. A hysteresis cycle is still observed when performing M-H measurements on the $x=1.4$ sample at room temperature, as awaited from the Curie temperature of 370 K for this composition. The observed saturation magnetization is of 90 emu/cm^3 and the coercive field of *ca.* 600 Oe at 300 K.

Pure *b*-axis growth of high crystalline quality GFO thin films has been obtained on two different conducting bottom electrodes : ITO (001) buffered YSZ (001) and Pt (111) buffered YSZ (111). The GFO thin films deposited on single crystalline Pt(111)/YSZ(111) electrodes exhibit excellent crystallization and the number of GFO in-plane variants is reduced from six to three. The measured leakage current on this latter electrode decreases by 4 orders of magnitude in comparison to the use of an ITO

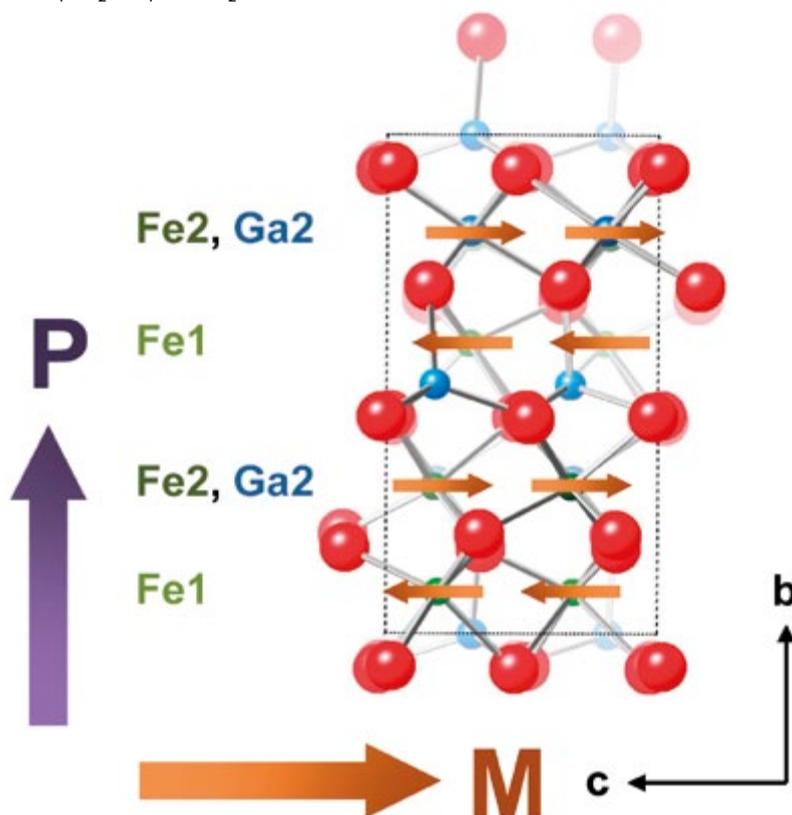
bottom electrode. Growth on a single crystalline Pt(111) electrode therefore leads to substantial improvement of both the crystalline quality and the dielectric properties of the GFO films.

This work opens new perspectives for the integration of a room temperature ferrimagnetic magnetoelectric material in spintronic devices. ■

Room temperature ferrimagnetic thin films of the magnetoelectric $Ga_xFe_{2-x}O_3$, M. Trassin, N. Viart, G. Versini, S. Barre, G. Pourroy, J. Lee, W. Jo, K. Dumesnil, C. Dufour and S. Robert, *J. Mater. Chem.* **2009**, 19, 8876-8880

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Crystallographic structure of GFO together with its net electric polarization (*P*) and magnetization (*M*) directions. The Ga^{3+} and Fe^{3+} cations are distributed on four types of sites labeled Fe_1 , Fe_2 , Ga_1 and Ga_2 . ▼



◆ Formation dynamics and nature of tryptophan's primary photoproduct in aqueous solution

Like other amino-acids tryptophan (Trp) is a building block of proteins. Spectroscopically though, it is very specific, since it is the brightest fluorophore among natural amino acids and its (UV) fluorescence emission spectrum and yield are strongly affected by its molecular environment. Therefore Trp fluorescence spectroscopy has long been established as a local probe for protein structure. For instance, the fluorescence spectrum strongly shifts depending on whether a given Trp residue is exposed to water or buried inside the protein. This has been used to address the issue of protein folding/unfolding. Alternatively, the fluorescence lifetime (or quenching) strongly depends on whether the electronic system of Trp is interacting with another one in a so-called pi-stacking interaction, an important mechanism for biomolecular recognition. However, the precise understanding of Trp photophysics remains far from trivial. Even when isolated in water, Trp fluorescence decay is multiexponential due to a confor-

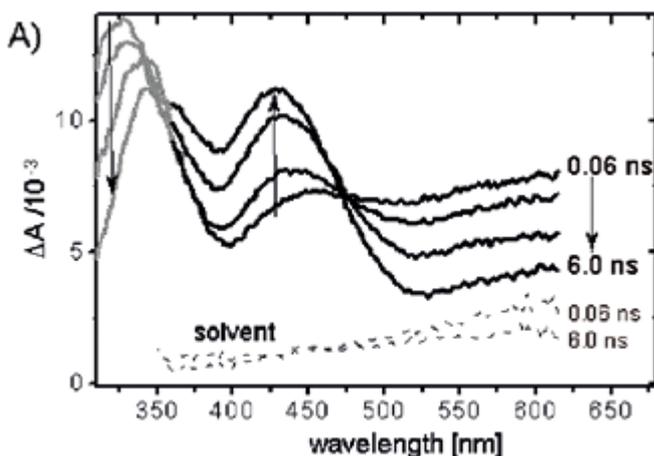
mation-dependent quenching mechanism debated over several decades. We have performed UV-VIS Transient Absorption (TA) spectroscopy in order to identify the formation dynamics and nature of the photoproduct resulting from S_1 quenching of Trp in water.

We discovered a new, spectral signature which is rising with the same multiexponential time dependence as what was previously observed for the fluorescence decay (see figure). Hence we conclude it is the absorption spectrum of the transient photoproduct resulting from excited state quenching. Like what was observed for fluorescence decay, the TA dynamics is sensitive to pH, and the time evolutions (photoproduct rise) exactly match what was reported by fluorescence spectroscopy for the excited-state decay. In addition we compare to the TA of Trp when it is incorporated in a protein, where we know that an excited-state electron transfer is responsible for the fast fluorescence quenching. In this case, we do not observe the above photoproduct signa-

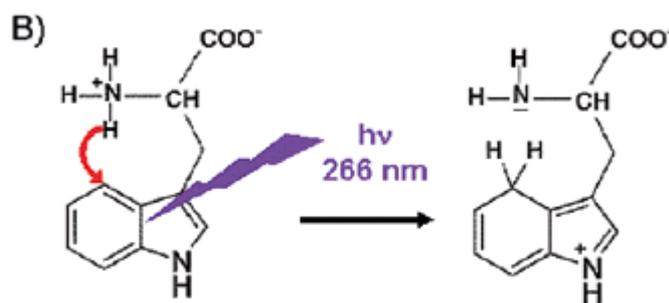
ture. Based on our observations and on previous works, we conclude that the quenching mechanism for Trp in water is due to a proton transfer rather than an electron transfer, thus concluding a 30 year-old debate. We also reveal the absorption spectrum and lifetime of this early photoproduct. Quantum chemistry modeling is now required in order to get deeper insight into the complex photophysics of Trp. ■

Formation dynamics and nature of tryptophan's primary photoproduct in aqueous solution, J. Léonard, D. Sharma, B. Szafarowicz, K. Torgasin and S. Haacke, Phys. Chem. Chem. Phys., 2010, 12, 15744-15750

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▲ Figure: A) Transient absorption change of Trp in Water, at pH=7, upon UV excitation at 266 nm. At early times (0.06 ns) we observe the excited-state absorption from the S_1 state of Trp, with a band around 325 nm and a weaker one around 450 nm. While time evolves up to 6 ns, this spectral signature decays (clearly seen around 325 nm and 500 to 600 nm) to give rise to a new spectrum characterized by two pronounced absorption bands at 350 nm and 425 nm attributed to the photoproduct.



▲ Figure: B) In water at neutral pH, Trp is in a zwitterionic form. Following UV light excitation, the dominant S_1 quenching mechanism is an intramolecular, excited-state proton transfer occurring on a few ns time scale.

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