Magnétisme et Chiralité

LNCMI, Grenoble

13-14 Juin 2023

Tuesday June 13th / Mardi 13 Juin

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14h45-15h05		Discussion on symmetries	Discussion sur l'apport de la symétrie	
15h05-15h25	Pause			
15h25-15h50	J. Long	Harnessing chiral lanthanide complexes for room temperature molecular ferroelectrics and magnetoelectric coupling	Exploitation de complexes de lanthanide chiraux pour la ferroélectricité moléculaire à température ambiante et le couplage magnétoélectrique	p. 3
15h50-16h15	V. Simonet	Spin texture in a chiral langasite: experiment and theory	Texture de spin dans une langasite chirale : expérience et théorie	p. 4
16h15-16h35		Discussion on magneto-electric coupling	Discussion sur le couplage magnéto-électrique	
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17h00-18h00		Visiting the facilities	Visites des installations	

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9h25-9h50	F. Pointillart	Magneto-Chiral Dichroism in Ytterbium Coordination Complexes	Dichroïsme Magneto-Chiral dans des complexes de coordination de l'Ytterbium	p. 7
9h50-10h15	B. Le Guennic	Magneto-Chiral Dichroism : a Quantum Chemist view.	Dichroïsme Magnéto-chiral: quel apport de la chimie quantique ?	p. 8
10h15-10h40		Discussion on MChD	Discussion sur le MChD	
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11h25-11h50	J. Crassous	Helicenes and Chiral-Induced Spin Selectivity	Helicènes et Sélectivité de Spin Induite par la Chiralité	p. 10
11h50-12h15	E. Pouget E. Hillard	Design of functional nanostructures via chirality induction	Conception de nanostructures fonctionelles via induction de chiralité	p. 11
12h15-12h45		Discussion	Discussion	
12h45-13h00	C. Train	Conclusion	Conclusion	

Magneto-chirality and symmetry

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Basically, chirality is the absence of mirror symmetry in a material or system, and magnetism corresponds to an absence of time reversal symmetry, two of the most fundamental symmetries in nature[1]. Each absence of symmetry leads to a class of specific physical phenomena. In the optical domain, chirality leads to the well-known optical activity and natural circular dichroism, whereas magnetism leads to the Faraday effect and magnetic circular dichroism. The simultaneous absence of both symmetries corresponds to a distinct case, which allows for the expression of a separate class of phenomena. This class is expressed in the optical domain in the form of magneto-chiral anisotropy, and in many other domains, like electrical and thermal transport, which only recently have been explored.

In this talk I will briefly discuss the underlying symmetry arguments and review existing and potential expressions of magneto-chiral effects[2].

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KEYWORDS: Chirality, Magnetism, Magneto-chiral anisotropy…

Second-Order Nonlinear Chiroptical/Magneto-Optical Techniques

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The detection of molecular and supramolecular chirality remains an important issue in a broad range of physical and chemical sciences. In practice, the detection and quantification of chirality is done with standard chiroptical methods such as optical rotation dispersion (ORD), circular dichroism (CD), circular polarization luminescence (CPL). Yet, a drawback of these methods lies in their rather low sensitivity (< 10^{-3}) such that chiral discrimination remains in some cases a challenging task.

In a first part, I will present recent results involving Inverse MagnetoChiral (IMCh) Birefringence in (a)chiral liquids, known as the Inverse Faraday effect (IFE) which is an all optical excitation process.[1] We have recently shown that, using a tunable picosecond laser source (700-2000 nm), Inverse Faraday effect (IFE) enables to obtain equivalent longitudinal magnetic flux densities in liquids in the range of 1-100 T with average optical pump power ranging typically between 100 µW to 10 mW. I will present an original all-optical method, that combines the inverse Faraday effect to measure the Faraday rotation angle (Faraday effect) and determine the Verdet constant of diamagnetic achiral and chiral liquid solutions. Finally, I will also emphasize the Verdet constant of chiral molecules exhibit large enantiomeric difference as at least +18.5% in the case of carvone at 775 nm and -10.0% in the case of α-pinene at 700 nm.

In a second part, I will present original and recent results obtained with hyper-Rayleigh scattering (HRS) in chiral liquids, a two-photon excitation scattering technique that offers quite different contrasts than classical chiroptical techniques. Hyper-Rayleigh scattering has long been predicted to be sensitive to the optical activity of chiral media. Recently, we have reported for the first time hyper-Rayleigh optical activity (HROA) from supramolecular organic systems, using volume less than 60µl.[2] I will show that any chiral liquid media exhibit large HROA contrast, for example as high than -6.3% for carvone and +17.3% for α-pinene. I will also show that HROA polarized responses, using only incident linear polarization, are characterized by symmetric and asymmetric terms which are, respectively, at the origin of the enantiomeric differences but also chiral optical activity differences, using +45° and -45° linear polarization excitation. So far, no theory is able to explain these differences.

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KEYWORDS: Nonlinear optics, Optomagnetism, Chirality, Inverse Faraday effect, Hyper-Rayleigh optical activity

Harnessing chiral lanthanide complexes for room temperature molecular ferroelectrics and magnetoelectric coupling

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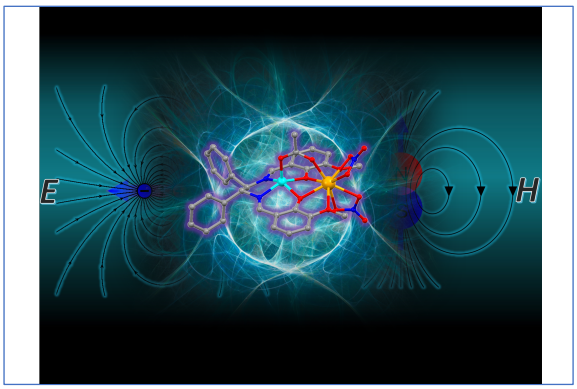
Coordination chemistry of lanthanide ions allows the tailored design of multifunctional molecule-based materials in which properties are gathered into a single molecular system. Yet, taking advantage of the multifunctional character in future applications will imply the presence of a strong interactions between the properties.

We describe the design of chiral lanthanide Schiff base complexes with a high degree of functionality, combining magnetism, optical activity, and lanthanide luminescence.



Remarkably, these molecular systems behave as ferroelectrics surpassing the Curie temperature of $BaTiO_3$ by 180 K, making them the highest temperature working molecular ferroelectric reported to date. Furthermore, we demonstrate the presence of a strong room temperature magnetoelectric coupling, resulting from the association between ferroelectricity and magneto-elastic interactions [1].

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KEYWORDS: Lanthanide ions, Ferroelectrics, Magnetism

Spin texture in a chiral langasite: experiment and theory

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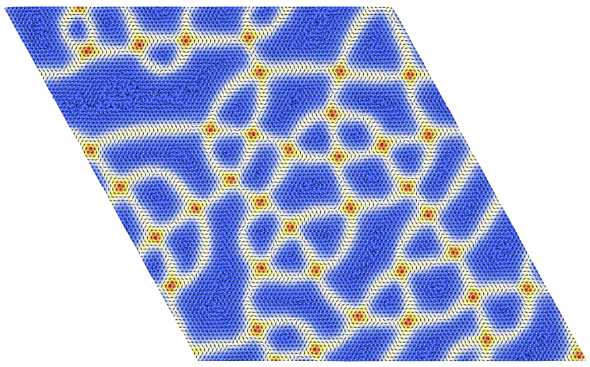
Iron langasites are model system that allowed to study structural and spin chirality and the relationship between them [1,2]. In this presentation, I will show the consequences of chirality on the multiferroic properties of this compound, associated to a very long wavelength spin texture, evidenced experimentally under magnetic field [3]. Il will also present a new theoretical study based on the minimal ingredients identified in these compounds, including magnetic frustration and Dzyaloshinskii-Moriya interaction, that predict novel spin textures and topological objects built upon the chirality vector [4].



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KEYWORDS: Chirality, Magnetism, topology

From high field MRI contrast agents to chirality detection – NMR studies in molecules at LNCMI

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Nuclear magnetic resonance (NMR) is a widely used technique for studying the static and dynamic properties of matter at the microscopic level. In addition to a long-standing NMR activity in solid state physics, the application of NMR at the LNCMI has recently been extended to molecular compounds. First, we developed a platform to characterize possible contrast agents for magnetic resonance imaging (MRI) based on lanthanide molecular magnets using ^{1}H-NMR up to ultra-high magnetic fields (33T / 1.4 GHz). Second, we started a first experimental attempt to directly detect chirality in molecules by NMR. For both projects, scientific goals, instrumental challenges and results are presented and discussed.

Magneto-Chiral Dichroism in Transition metals-based Molecules and Materials

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Magneto-Chiral Dichroism (MChD) is a fascinating but scarcely investigated manifestation of light-chiral matter interaction that manifests when chiral systems are placed in a magnetic field. It features an unbalanced absorption or emission of unpolarized light that depends on the relative orientation of applied magnetic field and light wavevector and the absolute chiral configuration of the system.[1,2] Its relevance is related to potential technological applications, such as the optical read-out of magnetic data, and its possible implication as a mechanism for the emergence of life homochirality.

With this talk I will provide an overview of the most recent results we have achieved on this topic, that are aimed at understanding the microscopic parameters and the chemical ingredients that are key to observe strong MChD responses. I will present the MChD observed up to ca. 40 K in two heterometallic chiral molecular ferrimagnets,[3,4] the key-role of spin-orbit coupling in driving MChD signals in a single-chain magnet based on tetragonally distorted Mn^{III} ions,[5] and the MChD response of a *3d-4f* chiral cluster made of Ni^{II} and Dy^{III} ions.[6]

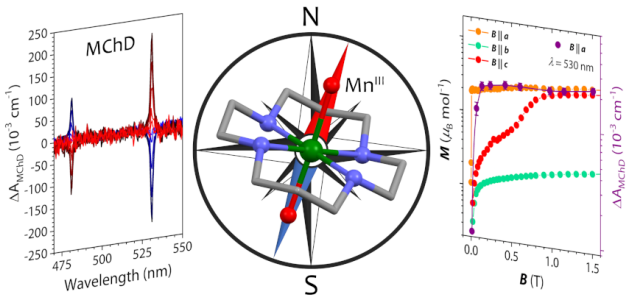

Figure: View of a portion of the molecular structure of one investigated Mn^{III} 1D chiral compound (middle), corresponding MChD signals observed by irradiation with visible light (left) and comparison between the intensity of optical and magnetic data.

Finally, I will present the first comparison between experimental MChD spectra and those theoretically calculated through *ab-initio* calculations, showing the fundamental role of vibronic coupling in enhancing the intensity and determining the shape of the MChD signals of chiral Ni^{II} complexes,[7] as well as further experimental results compared to calculated MChD spectra.[8]

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KEYWORDS: Chirality, Magnetism, Coordination Chemistry, Spectroscopy.

Magneto-Chiral Dichroism in Ytterbium Coordination Complexes

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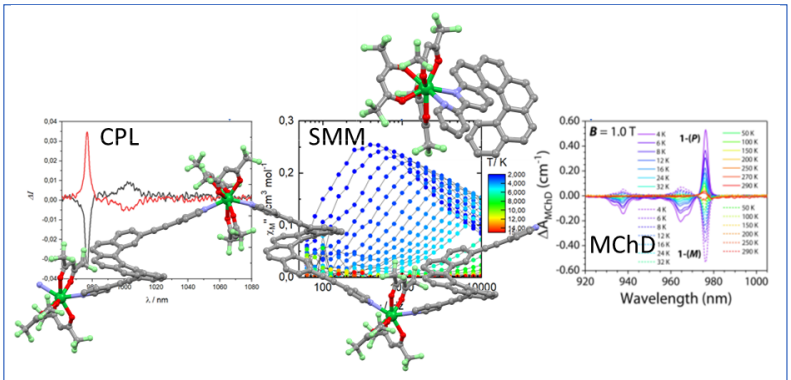
The design of non-centrosymmetric molecular magnets is of paramount importance since the absence of an inversion centre leads to efficient coupling of electric fields to the molecular magnetic moment,[1] and to magnetochirality,[2] a non-reciprocal effect that can be harnessed by unpolarised light. In other words, Magneto-Chiral Dichroism is the differential absorption or emission of unpolarized light that chiral magnetized systems exhibit under magnetic field. Such molecular magnets may combine magnetism with other physical properties displayed independently or in synergy leading to an attractive and unusual example of multifunctional materials merging chiroptical, luminescence, and magnetic properties.[3] Lanthanide ions are able to retain their magnetization in a given direction, thus generating a special class of Single-Molecule Magnet (SMM) thanks to their specific magnetic and optical properties.[4] In this context, we developed two new families of enantiopure Ytterbium complexes based on the inherently chiral helicene ligand[5]. Slow magnetic relaxation, Circularly Polarized Luminescence (CPL) and Magneto-Chiral Dichroism (MChD) were investigated for both mononuclear[6] and one-dimensional[7] compounds (Figure). In this presentation, the enhancement of the SMM performances and MChD effect in the polymeric structure will be argued thank to the support of ab initio calculations providing a deeper understanding of the underlying factors that govern the physical properties in these multifunctional nanomagnets.[7]



SMM), CPL and MChD behaviours observed in Ytterbium helicene complexes.

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KEYWORDS: Ytterbium, Helicene, Chirality, Circularly Polarized Luminescence, Magnetism, Magneto-chiral dichroism

Dichroïsme Magnéto-chiral: quel apport de la chimie quantique ?

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Au travers de quelques exemples issus de la littérature,[1] nous examinerons l'apport et les limites actuelles de la chimie quantique à la compréhension des mécanismes sous-jacents au dichroïsme magnéto-chiral. Pour finir, nous présenterons les efforts récents réalisés dans le but de reproduire et rationaliser les données MChD dans le cas spécifique des complexes de lanthanide.[2]

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KEYWORDS: Chirality, Magnetism, Quantum Chemistry

Chiral conducting materials for the eMChA effect

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Introduction of chirality into molecular precursors is a topic of much current interest as it allows the preparation of multifunctional materials in which the chirality may influence, for example, the conducting properties.[1] One of the strategies we have been developing over the last years consists in using chiral methylated BEDT-TTF and EDT-TTF derivatives in crystalline radical cation salts with diverse anions,[2,3] which allowed us, for example, to observe the electrical magnetochiral anisotropy effect (eMChA) for the first time in a TTF based conductor.[4] More recently, the two enantiomeric crystalline radical cation salts κ-[(*S,S*)-DM-BEDT-TTF]$_2$ClO$_4$ and κ-[(*R,R*)-DM-BEDT-TTF]$_2$ClO$_4$, showing κ-type arrangement of the organic layers, were investigated in search for superconducting chiral molecular materials.[5]

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KEYWORDS: Chirality, Conductivity, eMChA, Tetrathiafulvalene

Helicenes and Chiral-Induced Spin Selectivity

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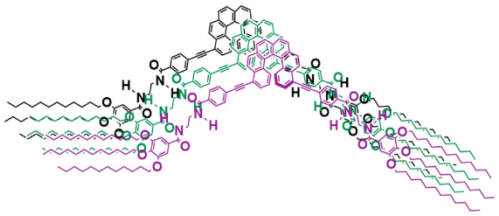
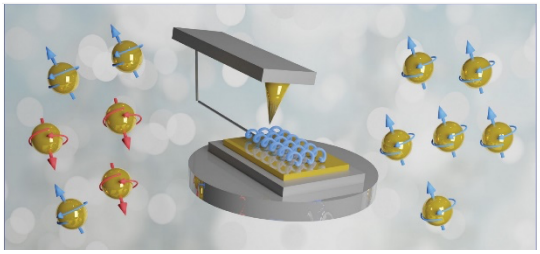
Ortho-fused aromatic rings form helically shaped chiral molecules such as carbo[6]helicenes, that wind in a left-handed (*M*) or a right-handed (*P*) sense.[1] The helical topology combined with extended π-conjugation provides helicenes with peculiar properties such as strong photophysical and chiroptical properties (high optical rotation values, intense electronic circular dichroism and circularly polarized emission). The molecular engineering of helicenes offers a convenient way to tune the properties of these helically shaped π-ligands. Indeed, their combination with metallo-organic units or organic aggregating substituents leads to chiral materials with appealing properties such as chiral-induced spin selectivity (the CISS effect).[2,3]





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KEYWORDS: Helicenes, Chirality, Spin selectivity, mC-AFM, Magnetoresistance, Electrochemistry

Design of functional nanostructures *via* chirality induction

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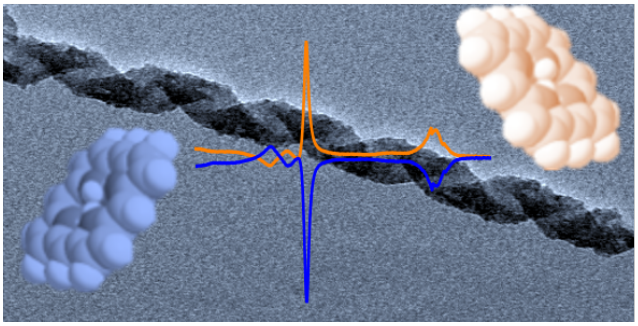
While the combination of chiral and magnetic properties is becoming more and more attractive for magneto-chiral phenomena, the fabrication of chiral systems that are highly responsive to both light and magnetic fields is still a challenge.[1,2] The induction of chirality to achiral objects is a versatile strategy worthy of exploration for the preparation of magneto-chiral objects. In this context, we investigate the use of silica nanohelices as a chiral platform for induced circular dichroism (ICD) to achiral magnetite nanoparticles, as well as achiral free base or metallized porphyrins.[3,4] These nanocomposites are studied principally by electronic natural circular dichroism (NCD) and magnetic circular dichroism (MCD) spectroscopies, and discussed as potential candidates for magnetochiral dichroism.



Electron microscopy image of TPPS porphyrin aggregated at the surface of a silica nanohelix and the CD spectra induced by the helices left and right.

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