Abstract book (Posters)

Asymmetric electron re-scattering in strong-field ionization of chiral molecules

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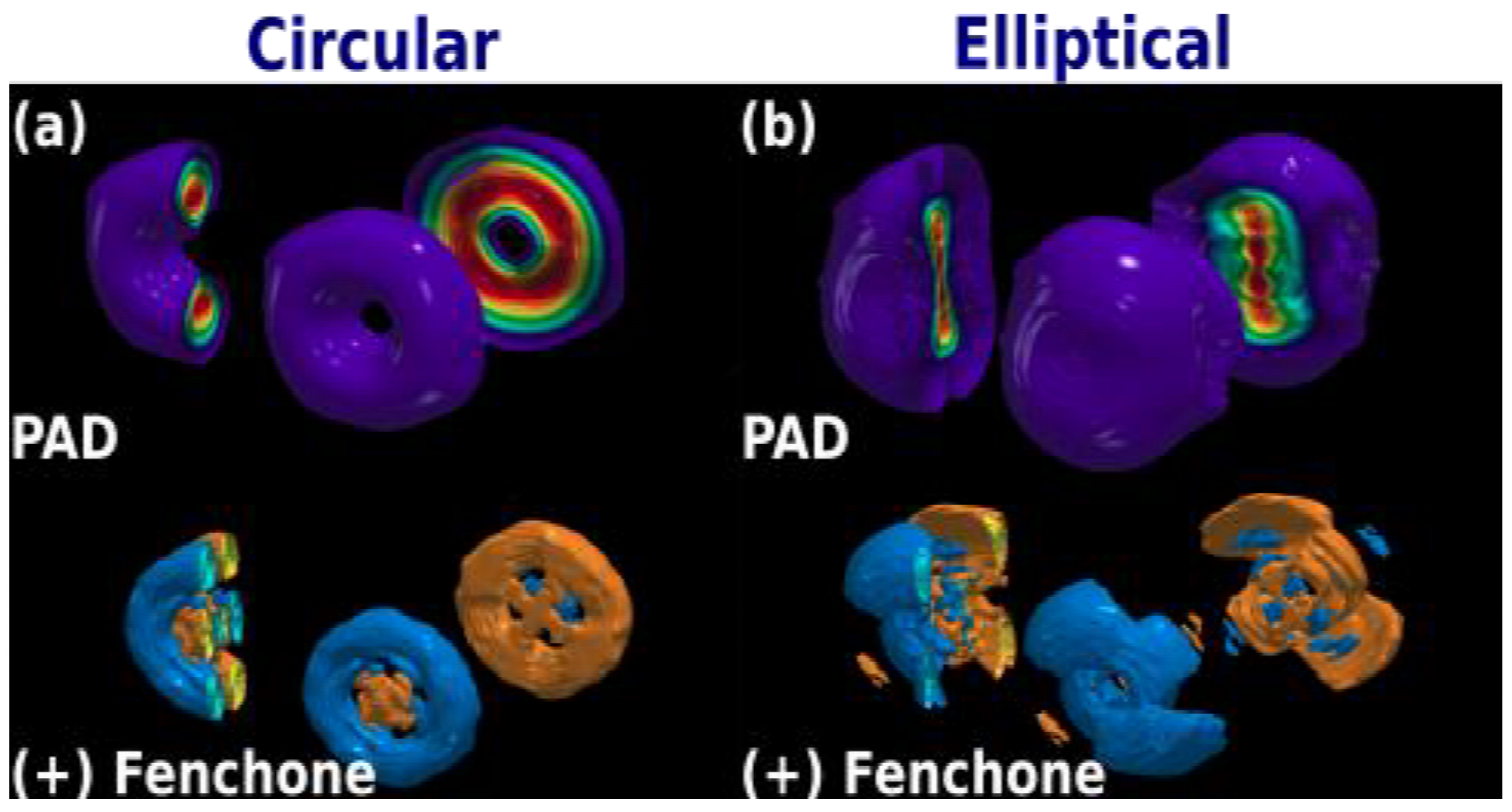
Abstract

When a molecule is ionized in the strong-field regime, an electron is born into the continuum by tunneling out of the lowered potential barrier formed by the electromagnetic field and the molecular potential. Depending on the time within the optical cycle at which the electron is released into the continuum, some of these electrons can be driven back by the electromagnetic field towards the ionic core and then re-scattered from the molecular potential before to reach the detector (indirect electrons). On the contrary some of them propagate directly towards the detector (direct electrons). The Photo-electron Angular Distribution recorded on the detector is thus a coherent superposition of these two types of trajectories and the information of the molecular potential is encoded through photo-electron holography or laser-induced electron diffraction(LIED). Retrieving the structural information for polyatomic molecules is rather challenging due to the complexity of the molecular potential and random distribution of the molecular orientation. In this work we propose a novel approach to retrieve these structural and temporal information using a symmetric property that survives random alignment of molecules: Photo-Electron Circular Dichroism (PECD).

Cuts of the 3-D PAD (up) and its forward-backward asymmetric component (down) is obtained by ionizing (+) fenchone molecules with a circularly (a) and a fixed elliptically (b) polarized laser field.



STAR-LIKE SELF-ASSEMBLIES OF CHIRAL FILAMENTOUS VIRUSES UNDER CONFINEMENT

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Abstract

Filamentous viruses have recently received growing attention as templates and building blocks in materials science. Due to their biological origin, they are highly monodisperse in size and shape making them a model system of colloidal chiral rods. When their repulsive interaction is turned into an attractive one by adding non-absorbing hydrophilic polymers, called depletion interaction, they self-organize into complex structures such as membranes, twisted ribbons and helical platelets. Here, we investigate the combined effects of depletion interaction and geometrical confinement in the self-assembly processes. We find that the rod suspensions exhibit a mesoscopic star-like morphology (Fig. 1) which could themselves self-organize into 2D lattices. Additionally, using optical microscopy techniques we determine the local ordering of the chiral viral rods within these structures.

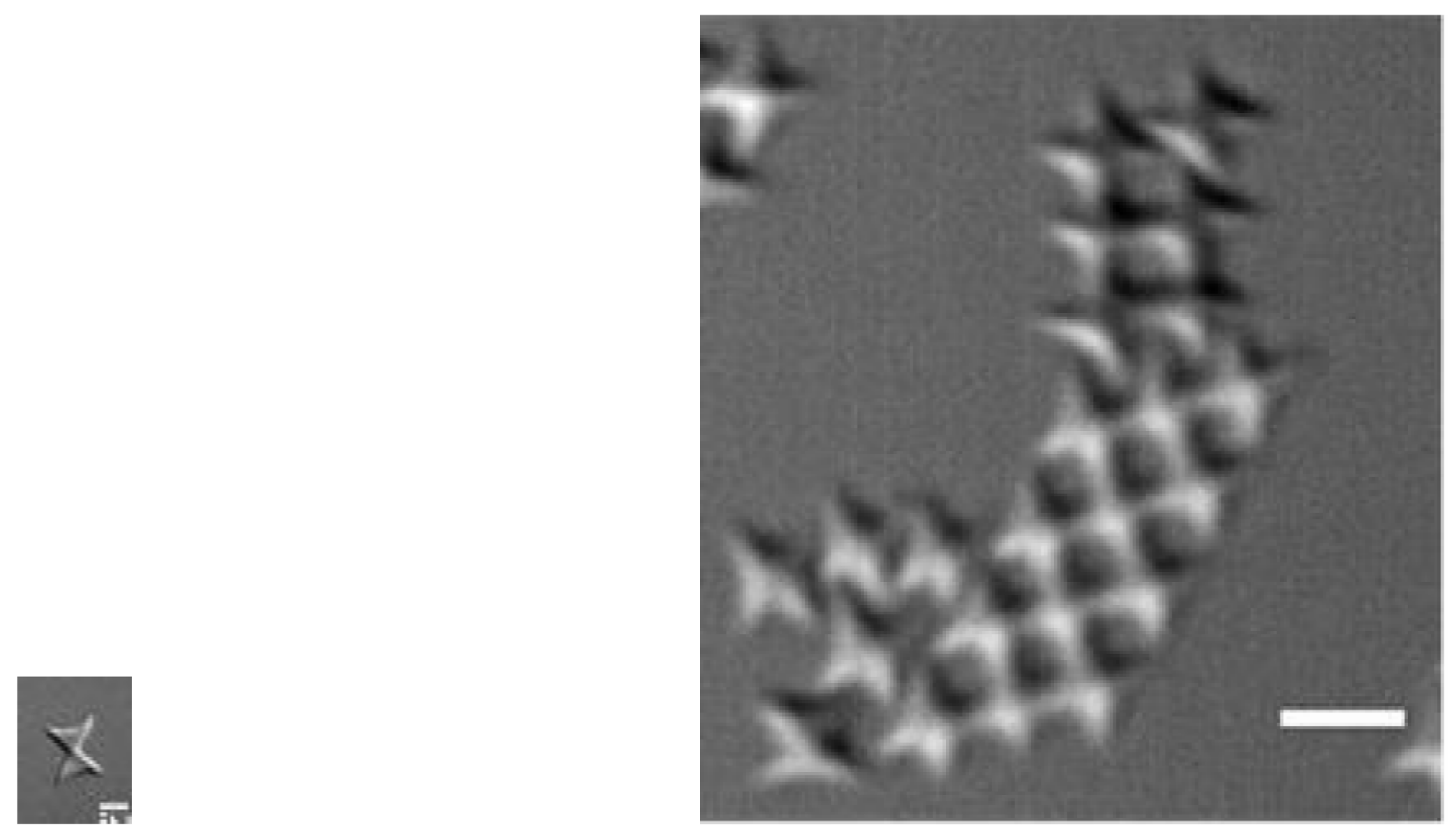

Fig. 1. Optical microscopy images of star-like **(left)** and lattice-like **(right)** morphologies

Chiral shaped perovskite nanocrystals synthesized via supersaturated recrystallization inside chiral porous silica ribbons

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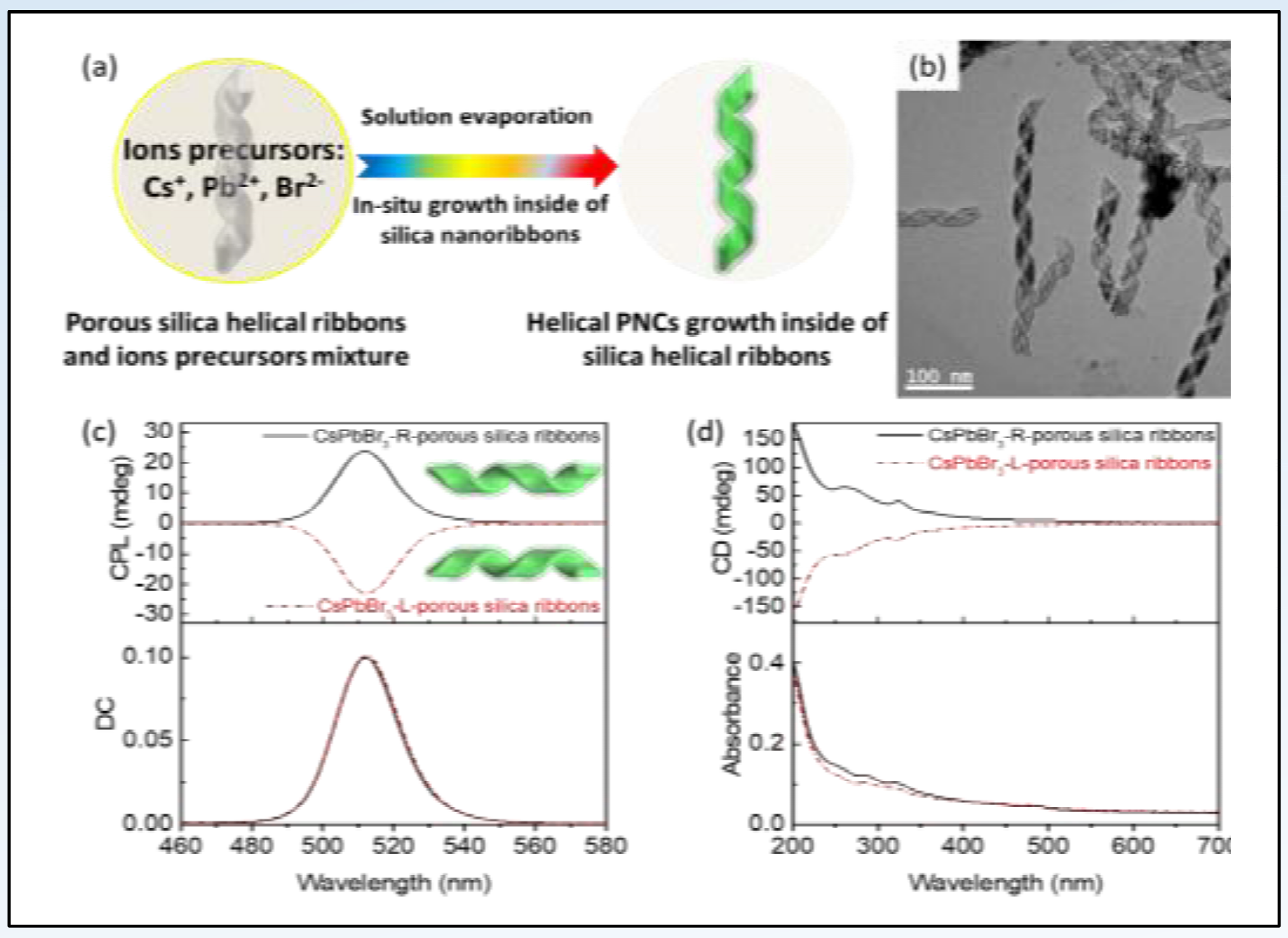

Abstract

Chiral perovskite nanocrystals (PNCs) have attracted lots of attention due to their excellent optoelectronic properties and promising applications in chiroptoelectronics, spintronics, and ferroelectrics. Recently, the chirality induced to PNCs by chiral ligands, and chiral organization has been widely reported. In here, PNCs having chiral shape with high dissymmetric g-factor are presented. These chiral PNCs are made using nanometric silica helical ribbons as platforms for the in-situ growth of the crystals using supersaturated recrystallization method. The PNCs grow inside chiral porous silica and result in a chiral shape as shown in figure 1 (a-b). The handednees of PNCs are determined by the handedness of porous silica templates. The right-handed PNCs show positive CPL signals, while the left-handed PNCs show negative CPL signals with dissymmetric g-factor up to $+/-2*10^{-2}$. Also, the right- and left-handed PNCs show strong mirror CD images which exhibit dissymmetric g-factor ($+/-1.5*10^{-2}$). Such a simple method to create inorganic materials having high dissymmetric g-factor is a very promising approach future applications.

MOLECULAR CHIRALITY ON A SHORT TIME-SCALE (fs-as)

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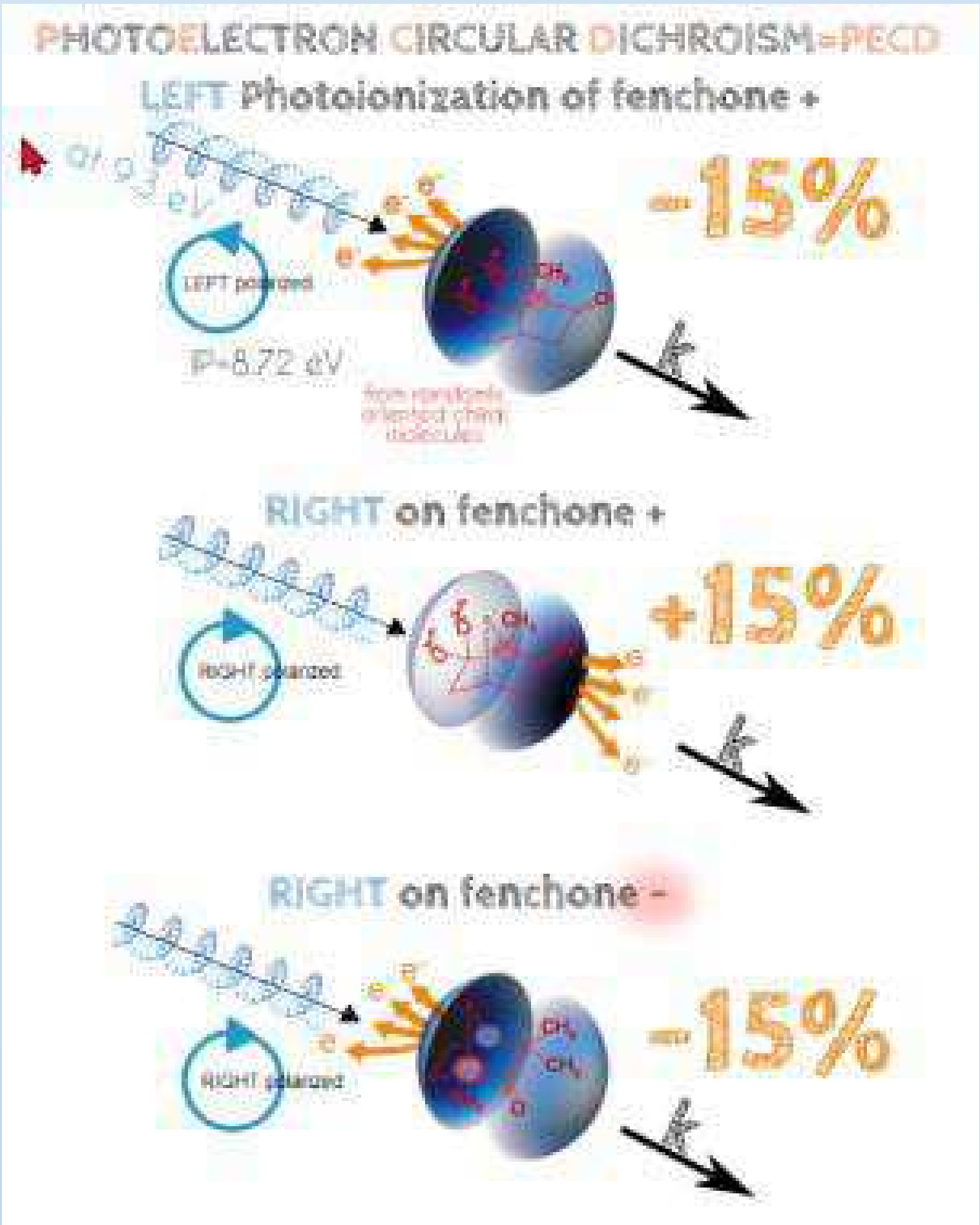
Abstract

Photoionization can produce strong chiral signals: when photoionization of chiral molecules is induced by circularly polarized light, the 3D-angular distribution of photoelectrons exhibits a forward/backward asymmetry with respect to the laser propagation axis. This effect has been called PhotoElectron Circular Dichroism (PECD) and results from the scattering of the electrons off the chiral molecular potential. It is quantum mechanically described as interferences between partial ionization waves. In this poster, different experimental approaches of time-resolved photoelectron asymmetry of chiral molecules will be presented ranging from a femtosecond resolution to attosecond one.

PECD in a nutshell: The asymmetry of photoelectron appears relative to the propagation axis of the photo-ionizing light. The asymmetry is said to be backward or forward depending of the light helicity and the enantiomer. Despite the fact that molecules are randomly oriented, the effect can reach an amplitude of several percents.



Enantioselective Brönsted acid-catalyzed additions of carbon nucleophiles to alkynes

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The carbophilic Lewis acid mode of activation of C-C bond unsaturation was developed during the two last decades as a new synthetic strategy for hydrofunctionalization reactions. Unfortunately, this carbophilic reactivity was observed for a set of late transition metals, including Au, Pt and Hg, characterized by their high cost and/or their relative low availability and/or their toxicity. Considering that the development of asymmetric transformations involving this type of activation would require additional chiral and often highly expensive ligands, the search for alternative original, catalytic and metal-free methodologies that could provide the same reactivity is of upmost interest.

Whereas the analogy of reactivity between carbophilic metallic Lewis and Brønsted acids (BA) is well established, the development of BA-catalyzed asymmetric additions of nucleophiles to alkynes is hampered by (*i*) the lack of reactivity of alkynes towards BA, (*ii*) the lack of polarisation of unactivated alkynes delivering a mixture of regioisomeric vinyl cations upon protonation; (*iii*) a lack of interaction between the carbocation and the chiral anion resulting in poor enantioinductions.

Our group postulated that the introduction of a suitable directing group (DG) on the alkyne could overcome these issues, and applications of this methodology to the synthesis of chiral atropisomeric phenanthrenes, alkenes and naphtalenes by Brönsted-acid catalysis will be discussed in this communication (*Adv. Synth. Catal.* **2019**, *361*, 2025).

Study of Chiral Recognition and Separation Processes by Silica Based Hybrid Materials

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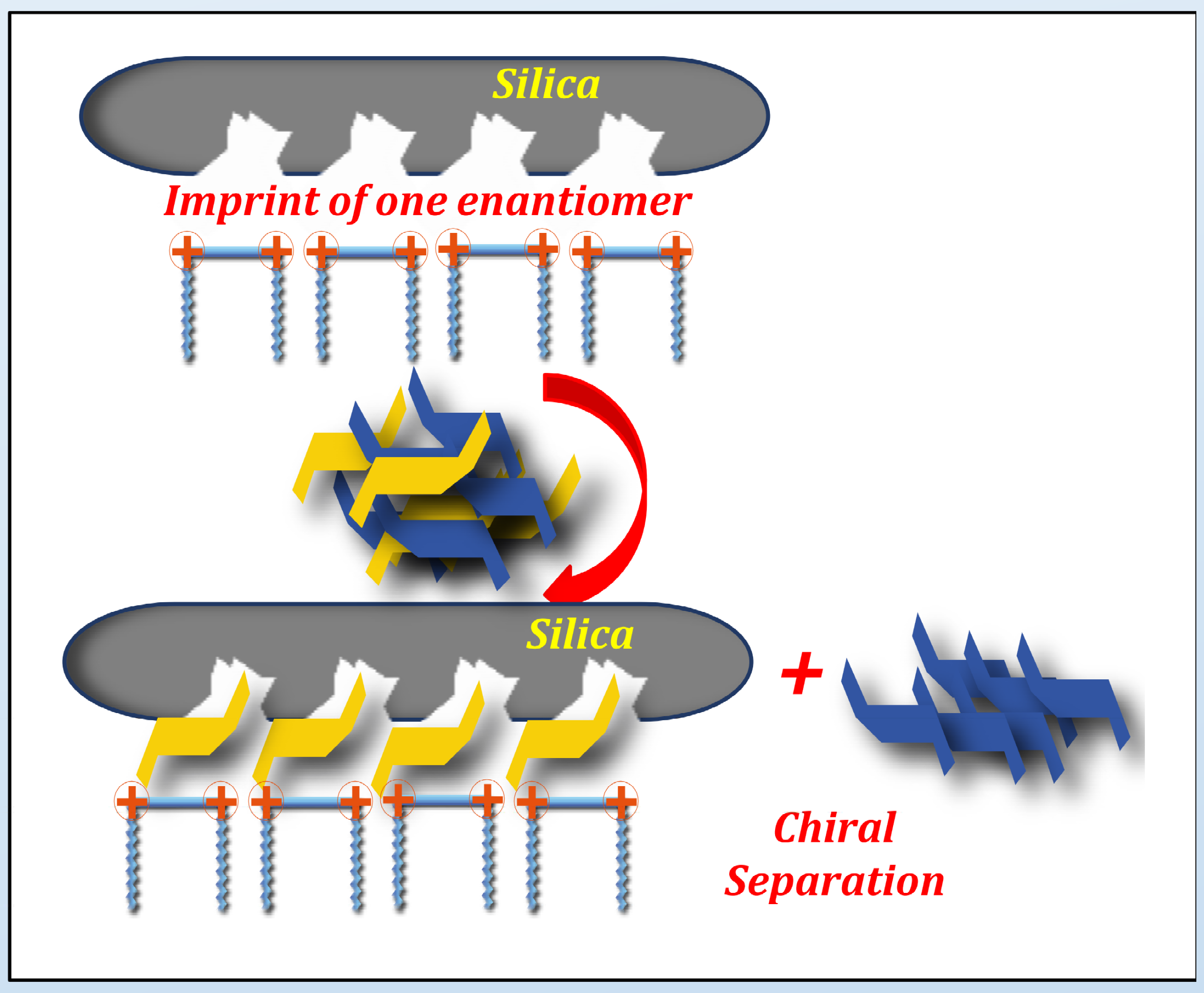
The transfer of chirality from a hybrid material such as nanohelices to achiral moieties has been reported majorly from our research group [1-3]. In the present study, the sensing and enantio-separation of relevant(pollutants) chiral molecules has been investigated by designed chiral hybrid materials. We have prepared our chiral selector with custom binding sites with memory of the shape, size and functional groups of the model molecules utilizing the molecular imprinting technology [4]. This method has is advantageous due to its low cost, ease of synthesis, high chemical and physical stability, excellent recyclability. Helical hybrid materials based on silica are considered due to its homochiral shape. We assess that such sensors can be applied for molecules similar in structure to the model molecules due to the facilitation of similar interactions. Handedness of the chiral selector controls the sensing or binding of the similar handed chiral analyte leaving the other enantiomer in the supernatant. This material can be applied as a filter for removal of chiral pollutants.



References: [1] N. Ryu et al., Chem. Commun.52 (2016) 5800. [2] J. H. Jung et al., ACS Nano 7(2013) 2595-2601 [3] J. Gao et al., ACS Nano 14(2020) 4111-4121. [4] S. Assavapanumat et al., J. Am. Chem. Soc. 141(2019) 18870-18876.

New chiral sensor-based porphyrin functionalized silica Nano-helices for enantioselective detection of chiral pollutants

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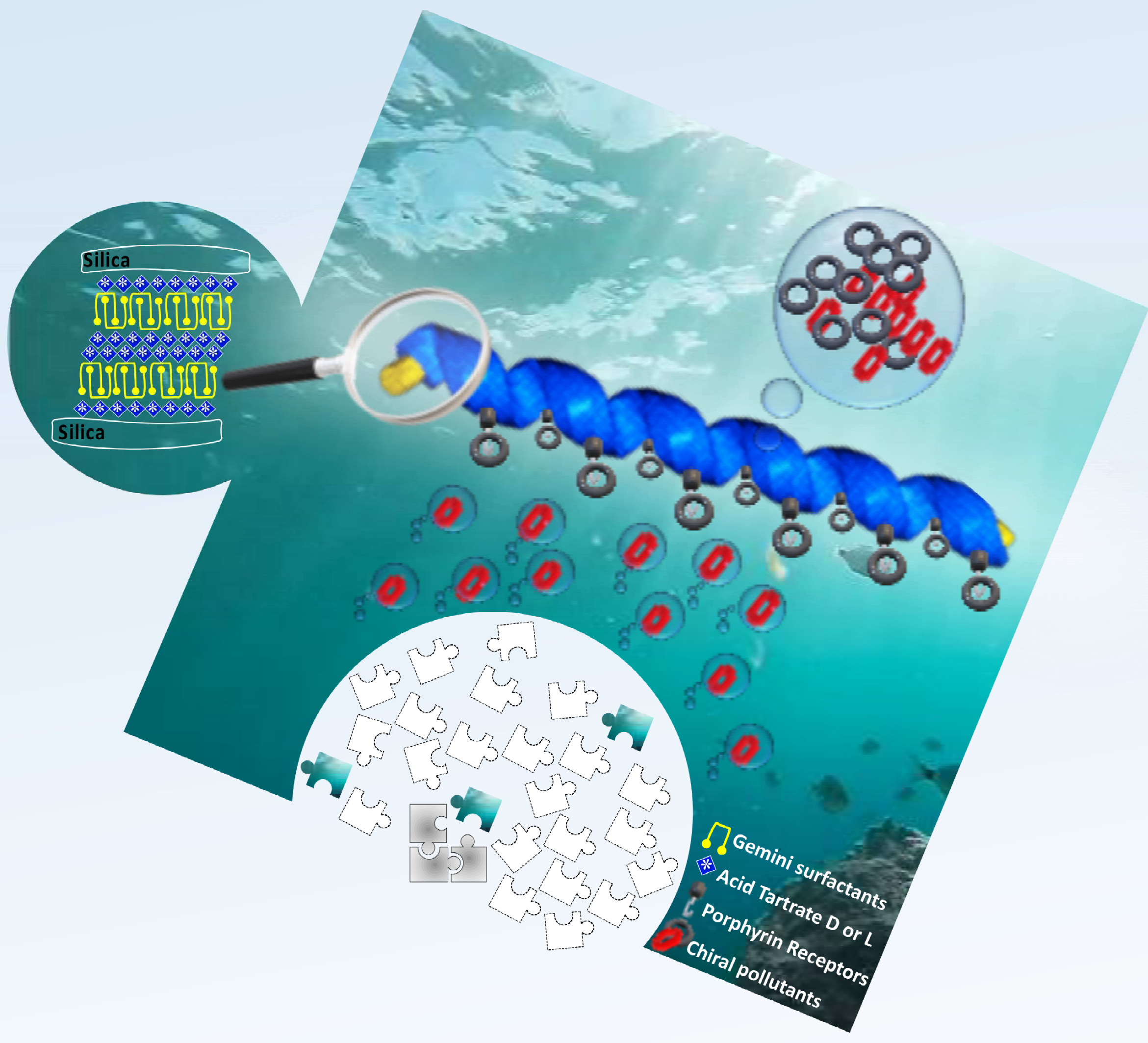
Abstract

Chiral pollution is a big threat to the human beings in terms of health, environment and economic losses. Therefore, discovery of new Nano chiral sensors for recognition of enantiomers is one of the most arduous challenges in chemical engineering and materials science [1]. Herein, chirality was induced to porphyrin derivatives molecules (which will be used as chiral sensors), which are complexed with silica Nano helices prepared as previously reported by our group (Fig. 1) [2-4]. The capability of such systems to behave as nanosensors to detect and recognize liquid or vapors of opposite enantiomers was evaluated by fabricating liquid/gas sensors based on quartz microbalances and by using electronic circular dichroism (CD). Chiral recognition and discriminations of chiral molecules through close charge-transfer, π-complexes or with Zn central atom will be discussed in the present work.



References:

[1] ACS Appl. Mater. Interfaces 2019, 11, 12, 12077–12087
[2] Jiaji Cheng et al (2017), ACS Nano. 11, 4, 3806–3818.
[3] Thomas delclos et al (2008), Nano Lett. 8, 7, 1929–1935.
[4] Reiko Oda et al (2008), J. Am. Chem. Soc. 130, 44, 14705–14712.

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Magnetic nanocomposites: toward magneto-chiral dichroism (MChD)

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Abstract

Magneto-chiral nano-objects are of great interest and could possibly lead to high-potential applications such as in spintronics or new magneto-optical devices.

Here functional nanocomposites are synthesized by using chiral silica nanohelices as a template for the grafting or deposition of magnetic species.

Helices can be modified to promote the grafting of magnetic compounds such as molecular complexes or superparamagnetic nanoparticles (SPM NPs). Our preliminary results have shown intense circular dichroism (CD) signals (particularly with the grafting of a heme analogue, see Fig. 1). These samples appear to be promising for the first observation of induced magneto-chiral dichroism.

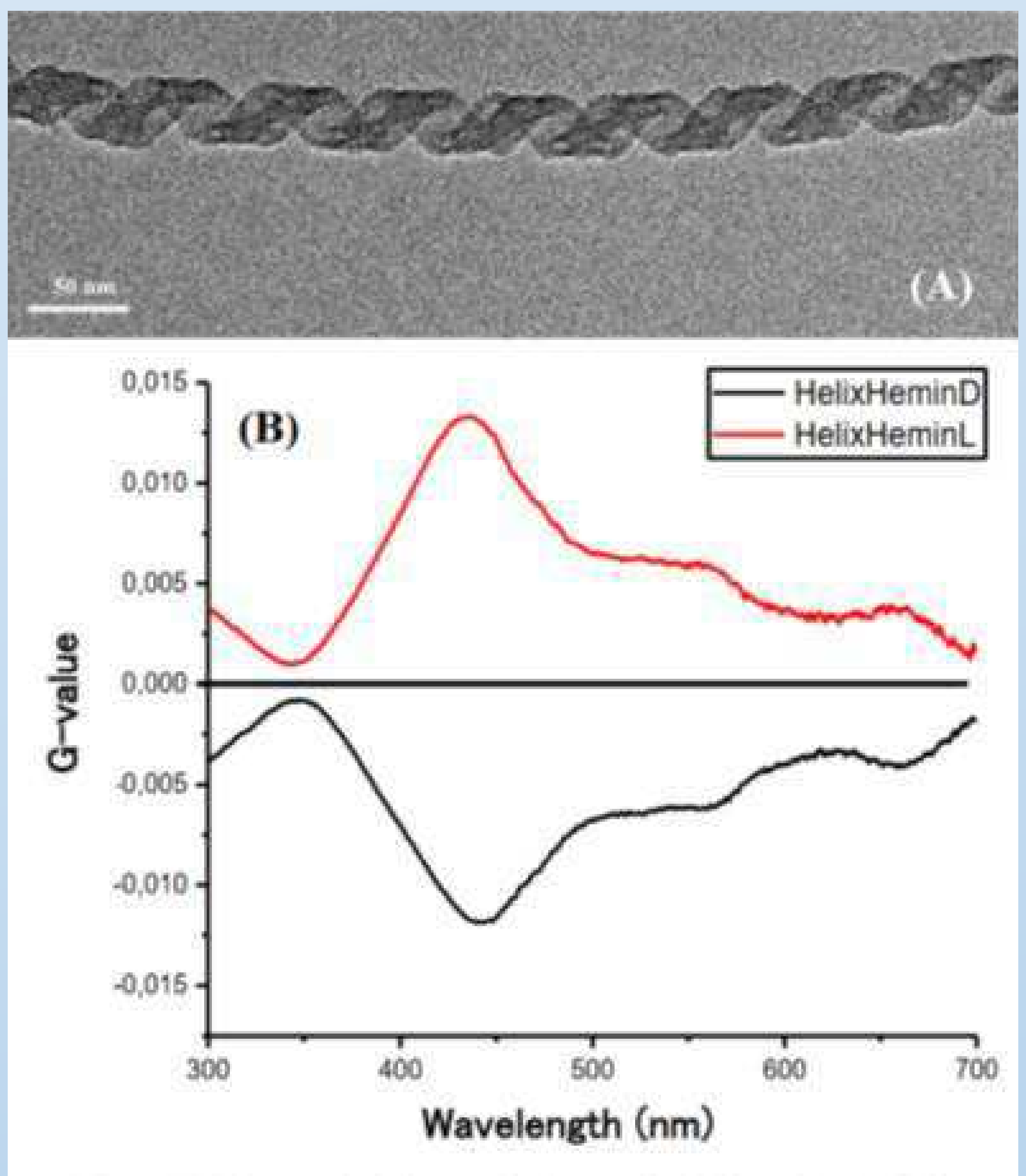

Fig. 1. TEM image of a silica nanohelix coated with hemin (A) and the corresponding CD spectra (B).

CHIROELECTROGENESIS: FROM STATIC TO DYNAMIC EVENTS

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Chirality is found throughout nature, but it occupies a special place in chemistry, perhaps for historical reasons, but mainly as a result of the beneficial properties of chiral molecules across a diverse range of areas, from medicine to materials science. In such scenario inherent chirality can provide the breakthrough. It implies chirality and key functional properties to originate from the same structural element endowing the selector with extraordinary chirality manifestations that can be propagated from molecular level to the macroscopic one. Unprecedented recognition, in terms of energy differences, was recently observed, implementing inherently chiral materials as enantiopure electrode surfaces. Recently, attractive potentialities of these systems were also exploited in the field of bipolar electrochemistry [1] and in the one of autonomous swimmers allowing to correlate the output signal with the concentration of the enantiomers present in solution. [2]

[1] S. Arnaboldi *et a.l Anal. Chem.* 92, 14, 10042–10047 (**2020**) ; S. Arnaboldi *et al. Chem. Mater*. 32, 24, 10663–10669 (**2020**)
[2] S. Arnaboldi *et al. Nature Chemistry* (**2021**) 10.1038/s41557-021-00798-9

Tunable Chiroptical Properties of Aromatic Oligoamide Foldamers

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Nowadays, there is a great interest in the development of new supramolecular systems with advantageous chiroptical properties that are circular dichroism [CD] and circularly polarized luminescence [CPL]. In this context, Aromatic Oligoamide Foldamers (AOFs) appeared as a suitable system to produce and modulate CPL emission. Indeed, AOFs are oligomers able to fold in helices with a high stability thanks to intramolecular interactions.[1] In these systems, chirality is expressed through the helical handedness which can be controlled by the introduction of a chiral inducer, to obtain *P*- and *M*-helices with an absolute selectivity.[2] Recently, a series of photoluminescent AOFs was synthesized to modulate the optical and chiroptical properties.[3] For this purpose, different achiral fluorophores were appended to aromatic foldamers to lead to strong chiroptical properties such as high luminescence dissymmetry factors (g_{lum}) by the order of $1.2x10^{-2}$ – $1.2x10^{-3}$. To go further, a new series of photoluminescent oligomers was designed and synthesized in order to bring new information and establish structure/chiroptical properties relationship.[4] Moreover, recent study demonstrates also interesting properties in nonlinear optics (NLO) field for this type of supramolecular system[5] in a manner to highlight the high capacity to control linear optics and NLO properties with AFOs.

[1] Huc, I. *J. Am. Chem. Soc.*, **2003**, 125, 3448–3449. [2] Huc, I., *J. Org. Chem.,* **2011**, 76, 195–200. [3] Olivier, C., *Chem. Commun.*, **2019**, 55, 9825–9828.[4] Olivier, C. *ChemPlusChem.,* **2021**, 86, 496–503.[5] Rodriguez. V. *J. Am. Chem. Soc.*, **2020**, 142, 257-263.

Chiral quasi-particles generated by chiral excitation

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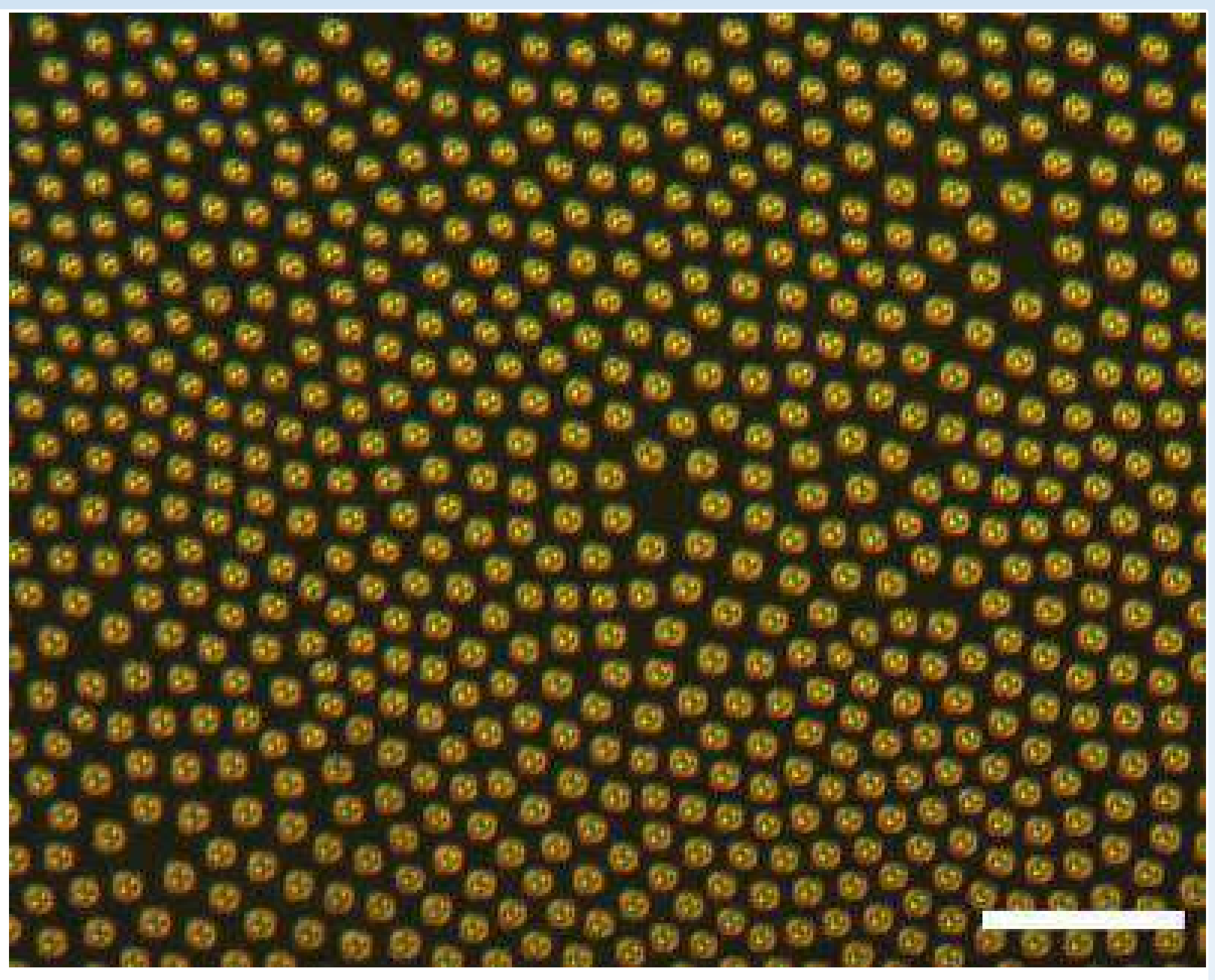
Abstract



Point-like structures in a frustrated cholesteric liquid crystal (100µm)

Liquid crystals are composed of molecules with positional disorder and orientational order. By adding a chiral dopant to rod like nematic phase, one can obtain a cholesteric liquid crystal. Imposing homeotropic boundary conditions to our sample leads to a frustrated cholesteric film, in which under certain conditions, localized elastic excitation may appear in various forms, for example finger-like patterns or more point-like structures.

These quasi-particles can be generated individually through the use of laser light, as an electric field applies a torque on the rod-like molecules, thus creating elastic excitation. The aim of my PhD is to master the generation of these quasi-particles as well as study how the characteristics of the excitation field can affect the properties of the generated objects. To perform this study, it is important to place ourselves in known frustration conditions to increase the robustness of our experimental approach independently of our sample characteristics. After this, we will look at the effect of incident field chirality (right or left circular polarization) on the structures, as preliminary studies have shown promising results towards polarization state recording in the material.

Magneto-Chiral studies on single crystals.

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Magneto-Chiral Dichroism (MChD) is a magneto-optical effect in which the absorption coefficient of a chiral molecule in an unpolarised light beam changes depending on the orientation of an externally applied magnetic field, parallel or antiparallel, to the direction of light propagation.[1] This phenomenon has been observed for the first time in 1997.[3]

In the present project, two well-characterized paramagnetic chiral materials, tris(ethylenediamine)nickel(II) nitrate and tris(ethylenediamine)cobalt(II) nitrate, have been selected for study (***Fig.1 A***). Enantiopure crystals were obtained by spontaneous resolution during crystallization. These crystals demonstrate very high MChD signals (***Fig. 1 B***), with *g* values up to 10^{-1}. Quantum chemical calculations have been developed, underlining the important contribution of vibronic coupling to the MChD intensity.[4]

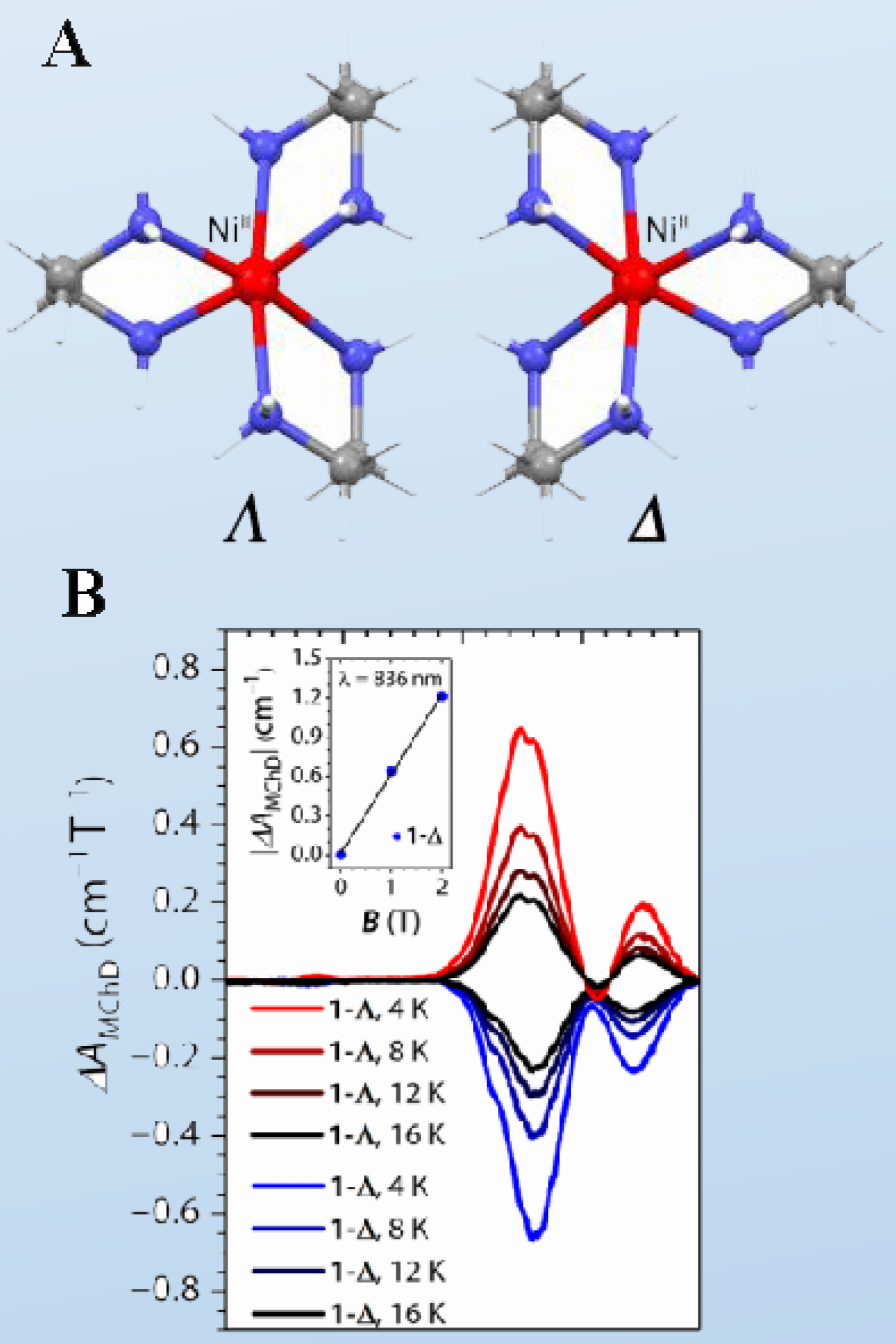

Figure 1. *(**A**) Molecular structure of the investigated systems. View of the molecular structure of Λ-$[MII(dae)^3]^{2+}$ (left) and Δ-$[MII(dae)^3]^{2+}$ (right) (MII = Ni, Co) complex cations.(**B**) Orthoaxial Δ_AMChD spectra for Λ and Δ single crystals for several temperatures.*

References: 1. M. Atzori, G. L. J. A. Rikken, C. Train, *Chem. Eur. J.*2020,*26*, 9784. 3. G. L. J. A. Rikken, E. Raupach, Nature 1997, 390, 493–494. 4. M. Atzori *et al.* Sci. Adv. 7, 2021.

Chiral Carbon dots obtained using silica nanohelices

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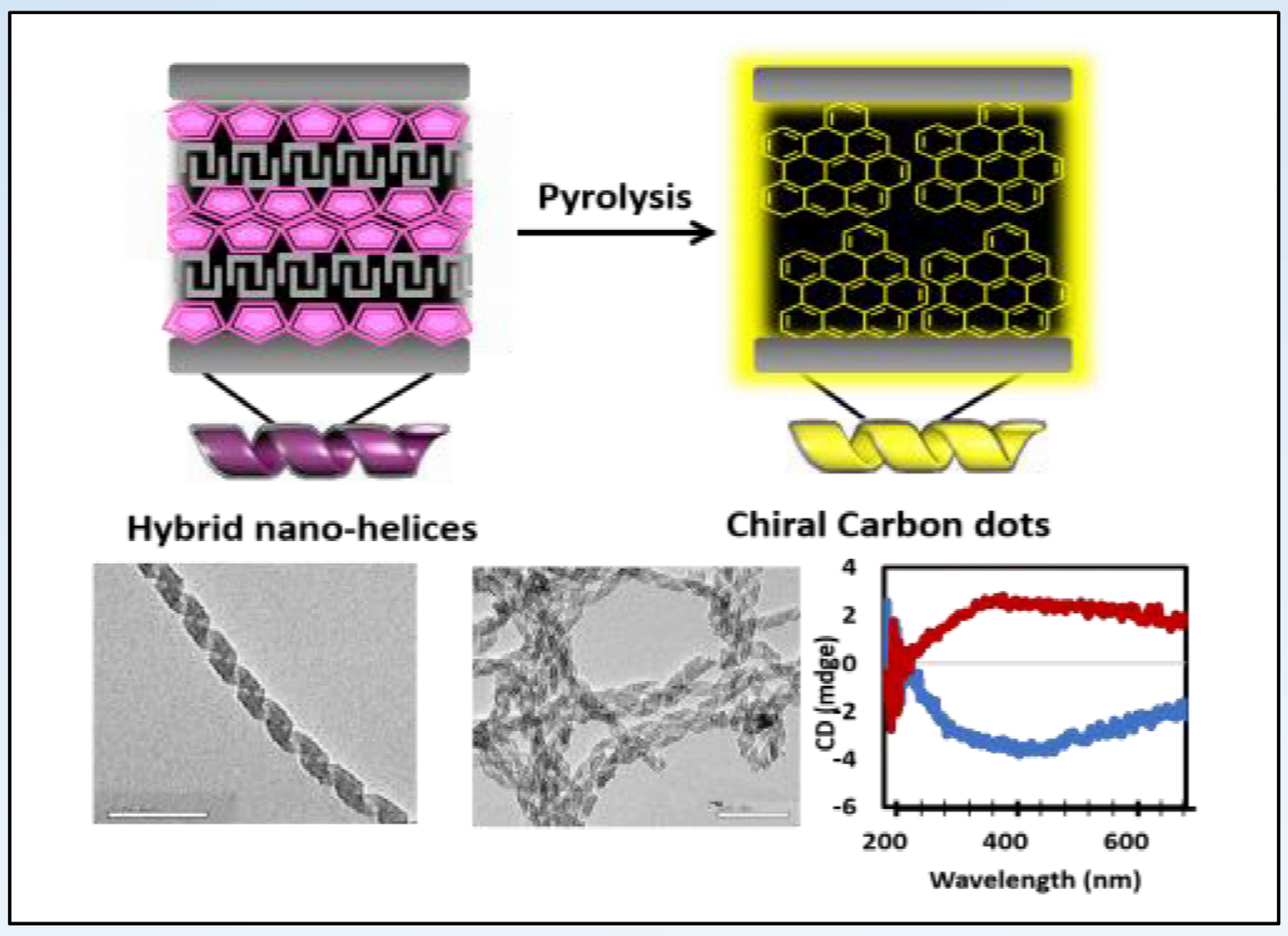
Abstract



One of the most interesting luminescent nanomaterials is carbon dots owing to their outstanding properties such as good biocompatibility, high stability and low toxicity [1]. Meanwhile, the chiroptical properties of luminescent materials have received a great deal of attention in the several areas such as photonics, pharmacology, nanomedicine and cell imaging [2]. Herein, the chiral carbon dots were fabricated via pyrolysis of organic molecules entrapped in left-handed or right-handed silica nanohelices. The chiral induction from nanometer-scaled to sub-atomic particles has been investigated. The supramolecular assemblies of gemini surfactant acts as a soft template to fabricate a helical silica shelf [3]. These silica nano-helices in turn keep the chiral property of the surfactant through their transformation to carbon dots during their pyrolysis. Additionally, the silica helices also allow stable dispersion of these carbon dots in various solvents. We expect that these chiral carbon dots have very promising potential for several applications in the future.

[1] Hola, K. et al (2014), Nano Today, 9 (5), 590-603.
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[3] Kazunori Sugiyasu, et al (2002), Chem. Commun., 1212-1213

New Chiral Iodanes: Synthesis, Structure Elucidation, and Evaluation in Asymmetric Synthesis

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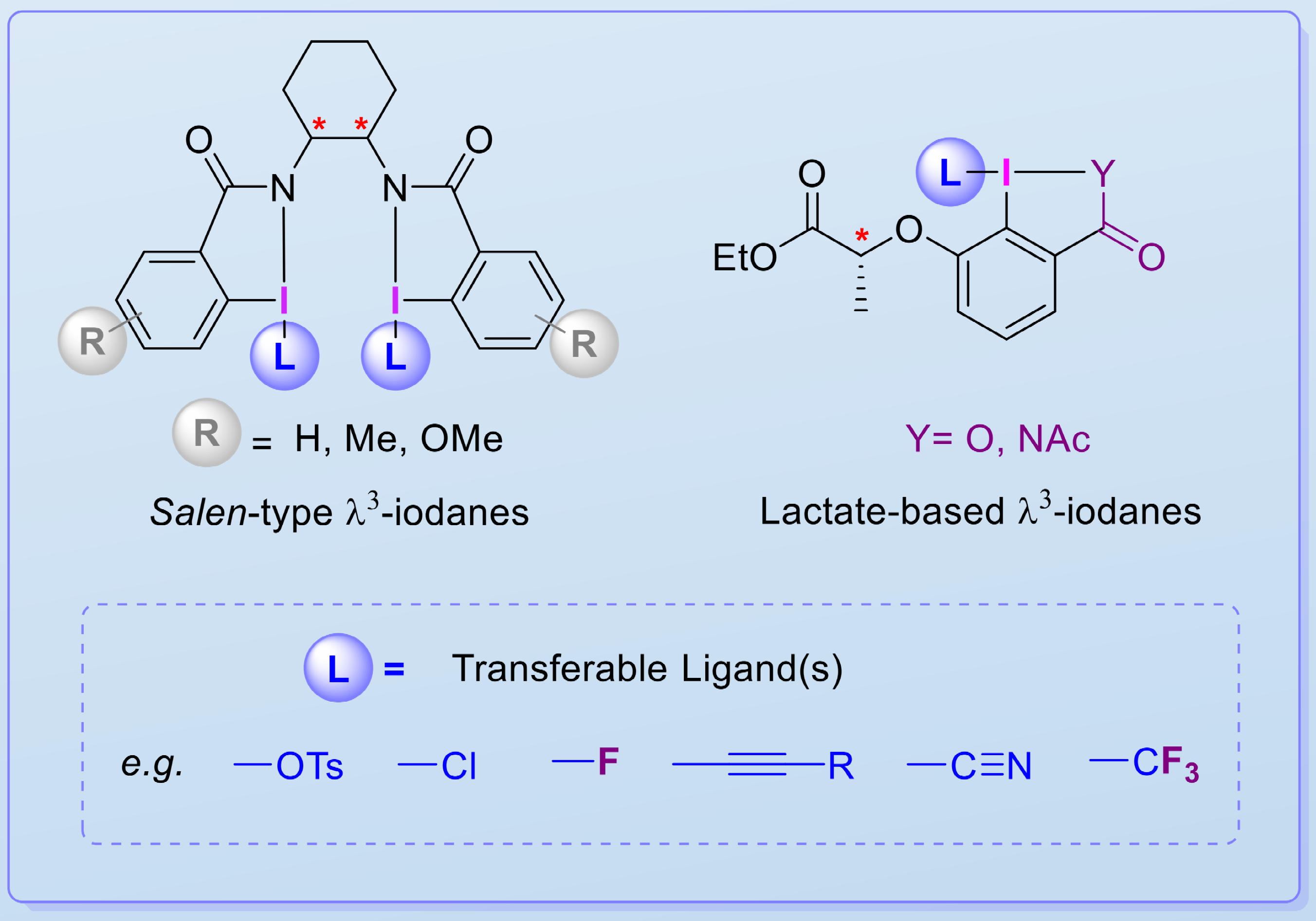
Abstract



Over the last decade, the search for **chiral iodoarenes** and **iodanes** has been one of the most competitive areas of investigations in **hypervalent iodine chemistry**, as these reagents represent a promising alternative to the use of (gradually rarefying) transition metals.[1] In this context, our group developed *inter alia* novel chiral *Salen*-type λ^5-iodanes and their use in stereoselective C–O bond-forming methodologies involved in natural product synthesis.[2],[3]
Herein, we present **new *Salen*-type** and **lactate-based chiral organo-λ^3-iodanes**, both featuring cyclic structures bearing heteroatomic or carbon-based transferable ligands. The synthesis and characterization of these iodine(III) reagents (*via* ^{13}C NMR and/or X-Ray analyses), and subsequent evaluation in asymmetric ligand-transfer reactions will be discussed.

Vibrational optical activity and induced ROA of solvent of chiral trinuclear paddlewheel complexes

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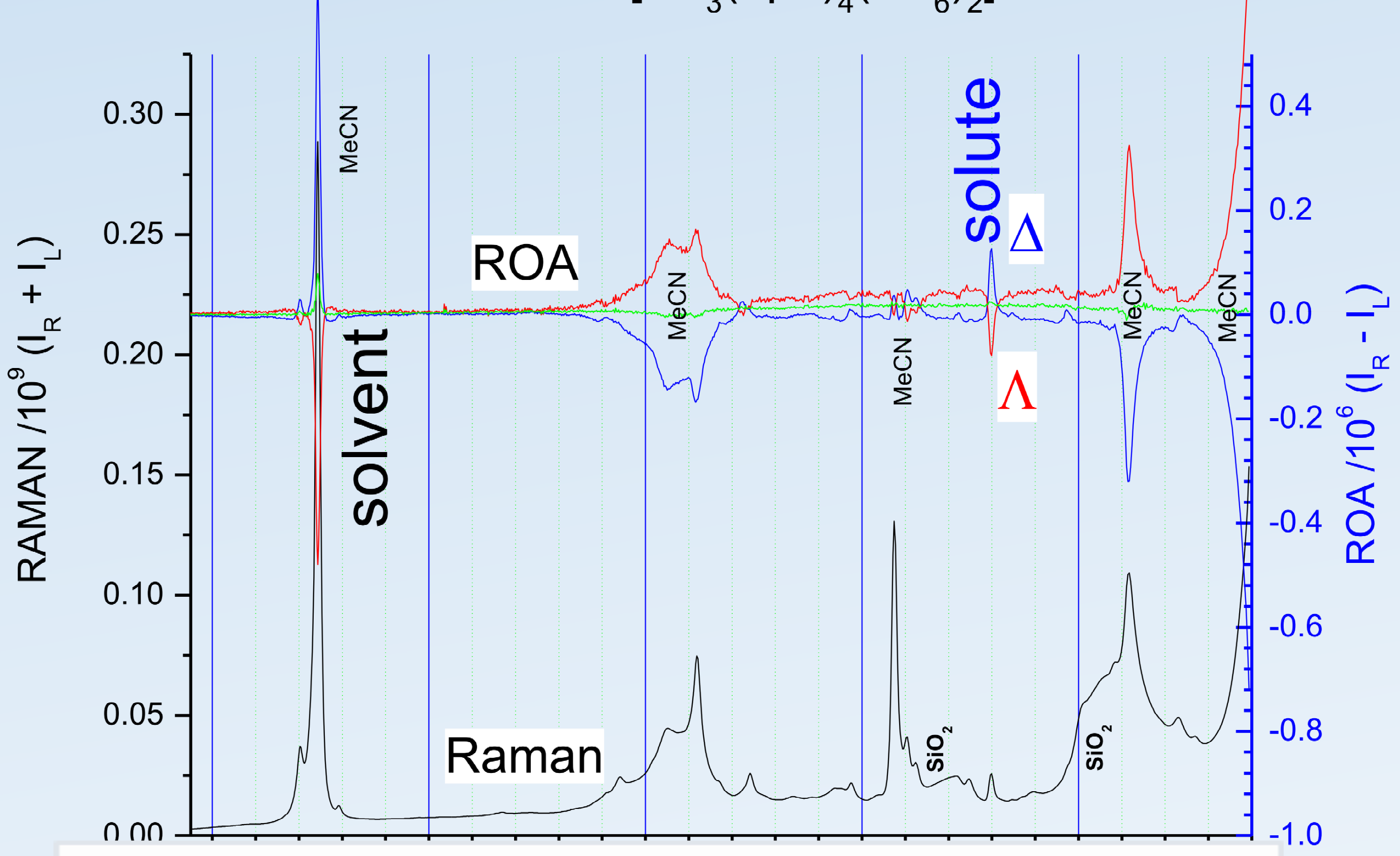
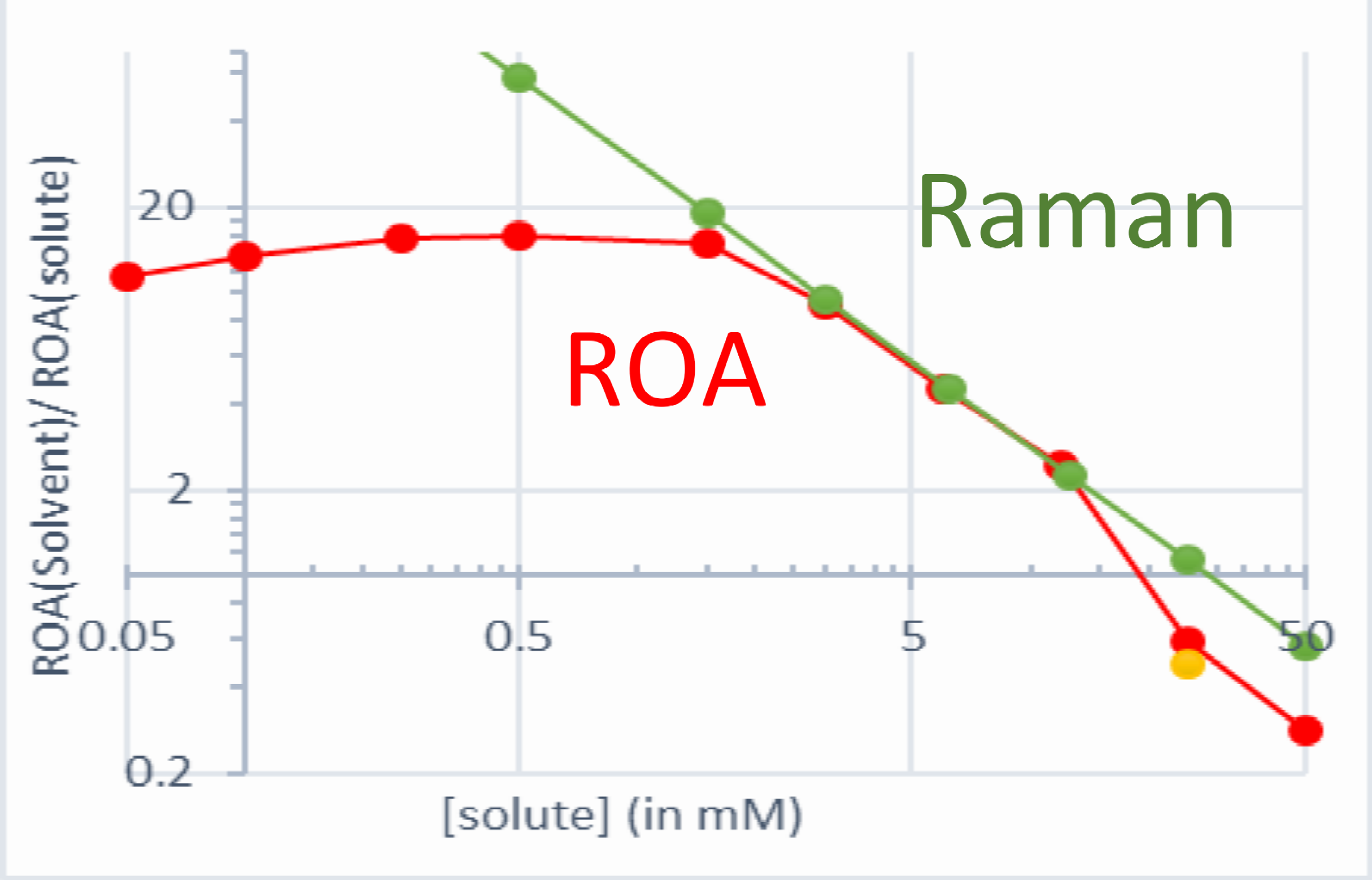
Abstract

Extended Metal Atom Chains (EMACs) are a fascinating class of linear polynuclear coordination complexes, which have served as a rich playing field for the study of metal-metal bonding, magnetism and electrical conductivity [1]. In addition, most trinuclear EMACs demonstrate helicoidal chirality arising from steric hindrance between the protons in the 3-position of the pyridine groups in the dipyridylamine (dpa) ligand. These compounds exhibits high specific optical rotation (> 5000 deg.mL.g^{-1}.dm^{-1}), interesting magnetic properties and unambiguous optical activity spectra (X-ray natural circular dichroism and ECD) [2].

In this study, we will focus on the intense vibrational optical activity signatures of the tri-metallic complexes Δ- or Λ-$[M_3(dpa)_4(MeCN)_2]^{2+}$ (M = Co, Ni). As already reported in the literature for helquat dye [3], the ROA spectra reveal a marked chirality induction in acetonitrile solvent, which is not observed on VCD spectra. We will discuss this strong induced solvent ROA and in particular the effect of the dilution (figure right) and its recent interpretations [3-5].

6 mM of Δ- and Λ-$[Co_3(dpa)_4(PF_6)_2]$ in MeCN





$$\frac{Isolvent}{Isolute} = f([solute])$$

(top) Model of Δ and Λ-$[Co_3(dpa)_4(MeCN)_2]^{2+}$, ROA (red and blue), Raman (black), solvent is MeCN
(bottom) Dilution study of the ratio $I_{solvent}/I_{solute}$, ROA (red), Raman (green)

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[4] Li, G. and al, *Angew. Chem.*, Int. Ed. 58, 46, 16495– 16498 (2019)
[5] Wu and al, *Angew. Chem.*, Int. Ed. 59, 49, 21895–21898 (2020)

Fast, accurate and universal measurement of the enantiomeric excess based on photoelectron elliptical dichroism

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Abstract

Photoelectron elliptical dichroism (PEELD) arises when a strong laser field elliptically polarized ionize a chiral chemical compound. The electrons emitted during ionization show a strong forward/backward asymmetry (1-10%) along the laser propagation axis, that reverse sign if the helicity of the light or of the molecule is changed.

With a high repetition laser source, the PEELD has already shown that it could determine the enantiomeric excess with an accuracy of 0.4% in 10 min acquisition time. The main advantage of the PEELD technology is that it is not a separative method that is inherently not universal and slow while keeping an excellent precision and accuracy compared to other optical methods.

However, the complexity of the technology makes its viability questionable. We solve this issue by presenting a demonstrator using a 10x less powerful laser, coupled to an 8x smaller photoelectron spectrometer compare to the initial experimental setup. It is able to determine the enantiomeric excess of a set of molecules, with excellent precision and great ease of use.

Picture of the demonstrator