Résumés

ELLIPSOMETRIE FEMTOSECONDE :

Aspects techniques et applications

Le 9 décembre 2019
Au FIAP, 30 rue Cabanis, 75014 Paris

Lundi 9 décembre 2019

09:15 - 09:45	*Accueil / Café*
09:45 - 10:30	Chiralité et Dichroïsme circulaire vibrationnel - Thierry Buffeteau
10:30 - 11:15	Dichroïsme circulaire de photoélectron - Yann Mairesse
11:15 - 11:30	*Pause café*
11:30 - 12:15	Dichroïsme circulaire électronique femtoseconde - Malte Oppermann
12:15 - 13:30	*Déjeuner*
13:30 - 14:15	Imagerie Polarimétrique - Marie-Claire Schanne-Klein
14:15 - 15:00	Femtomagnétisme - Nicolas Bergeard
15:00 - 15:15	*Pause café*
15:15 - 16:00	Faisceaux lumineux à moment orbital angulaire - Etienne Brasselet
16:00 - 16:30	Discussions - Bilan et perspectives utilisateurs

Dichroïsme Circulaire Vibrationnel (VCD) : origine, mesure et exemples d'application

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L'origine du dichroïsme circulaire vibrationnel (VCD) et ses particularités par rapport à l'absorption infrarouge conventionnelle seront présentées au cours de cet exposé. Après avoir défini les grandeurs pertinentes, nous verrons comment le signal VCD peut être mesuré expérimentalement et comment les spectres VCD peuvent être calculés à partir de calculs de chimie quantique. Quelques exemples d'application du VCD seront ensuite discutés : la détermination de la configuration et de la conformation absolue de molécules chirales, la mise en évidence de la chiralité dans des systèmes moléculaires organisés, l'utilisation du VCD pour identifier et quantifier les structures secondaires de polypeptides ou de protéines en solution ou déposés en films minces ...

Femtosecond and attosecond dynamics probed by photoelectron circular dichroism

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When randomly oriented chiral molecules are ionized by cicularly polarized light, the photoelectrons are preferentially ejected forward or backward the light propagation axis, depending on the handedness of the molecule and light. This chiroptical process, called photoelectron circular dichroism, is remarkably intense and sensitive. I will show how it can be used to track femtosecond dynamics of chiral molecules, as well as how the attosecond dynamics underlying the chiral photoionization process can be measured.

Capturing the ultrafast evolution of excited state chirality with time-resolved electronic circular dichroism

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For understanding the functioning of many photo-driven molecular machines, it is crucial to capture the temporal evolution of their chirality and thus structure upon photo-excitation. This is especially relevant for understanding many biological systems, such as photo-active proteins and the kinetics of novel synthetic machines, such as unidirectional molecular motors. Steady-state electronic circular dichroism (CD) has become a standard analytical tool to obtain solution-phase structural information via a molecule's chiral properties. Especially in the deep ultraviolet (UV) spectral range (< 300 nm), it is sensitive to the UV-transitions in proteins and many organic ligands in functional chiral complexes. However, pushing CD spectroscopy into the time-domain has remained a challenge, with only few isolated reports with sub-nanosecond resolution [1].

In this context, I will present a novel broadband time-resolved CD spectrometer in the deep-UV region (250–370 nm) with sub-picosecond time-resolution [2]. With this instrument, it is possible to extract broadband CD spectra of photo-excited states and track their temporal evolution. From the characterisation of the structurally well-defined photo-excited triplet metal-to-ligand charge transfer (MLCT) state in $[Ru(bpy)_3]^{2+}$, I will proceed to the more complicated dynamics of photo-excited supramolecular systems: $[Fe(bpy)_3]^{2+}$ is configurationally labile in solution and its configuration needs to be controlled by supramolecular complex formation with enantiopure counterions [3]. Here, we study the transfer of the underlying diastereomeric interaction to the photo-excited neutral quintet state, which couples to an accompanying ultrafast expansion of the metal-ligand distances.

I will conclude the talk by introducing a site-specific CD-label for time-resolved studies of ultrafast conformational dynamics in peptides and proteins [4]. In this way I hope to demonstrate how the measurement of broadband excited state CD spectra can open novel avenues in chiroptical spectroscopy and structural dynamics research.

[1] J. Meyer-Ilse et al., Laser Photon. Rev. **7**, 495 (2013)

[2] M. Oppermann et al., Optica **6**, 56 (2019)

[3] J. Lacour et al., Angew. Chem. Int. Ed. **37**, 2379 (1998)

[4] M. Oppermann & J. Spekowius et al., J. Phys. Chem. Lett. **10**, 2700 (2019)

Second Harmonic imaging of collagen 3D architecture in biological tissues: fast linear polarization resolution and Circular Dichroism

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Collagen is the main component of connective tissues such as arteries, skin, bones or cornea. It is mainly organized as fibrils of various sizes and 3D distributions depending on the tissue. A defective collagen 3D organization or synthesis can lead to pathologies or tissue malfunctions, including poor mechanical behavior. *In situ* imaging of these 3D structures is therefore a major biomedical concern.

The gold standard technique for *in situ* 3D visualization of collagen fibrils in intact tissues is second harmonic generation (SHG) microscopy. Polarization-resolved SHG microscopy provides further information about the hierarchical organization of collagen, from the molecular structure to the mean orientation and degree of alignment of collagen fibrils at micrometer scale. In this work, a fast polarization-resolved SHG microscope is implemented using line-to-line switching of polarization by an electro-optical modulator and working in epidetection geometry. It is fast enough to obtain reliable orientations in dynamic collagen samples, while slow enough to get a good SHG signal to noise ratio in thick collagenous tissues. We demonstrate that it enables accurate quantitation of dynamic collagen reorganization in murine skin dermis during stretching mechanical assays.

Moreover, Circular Dichroism SHG (CD-SHG) microscopy is implemented to take advantage of collagen chirality to improve 3D visualization. It measures the normalized difference in the SHG signal obtained upon excitation by left versus right circular polarizations. In this work, we identify two major artifacts that may occur in CD-SHG experiments and we demonstrate that thorough optimization and calibration of the experimental setup are required for CD-SHG imaging. Finally, we successfully record CD-SHG images in human cornea sections and confirm that this technique efficiently reveals collagen fibrils oriented out of the focal plane.

Laser induced ultrafast spin dynamics in rare-earth / transition metals alloys by element- and time-resolved X-ray Magnetic Circular Dichroism

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Femtomagnetism is one of the hottest topic in modern magnetism ever since its discovery in 1996 [BEA96]. The numerous theoretical and experimental works in this field of research aimed at identifying the microscopic mechanisms that allow the transfer of the angular momentum away from the spin degree of freedom during the laser induced ultrafast demagnetization. In this context, my main scientific interest concerns the investigation of laser induced ultrafast spin dynamics in rare-earth (RE) / transition metals (TM) alloys. Indeed, these alloys offers unique opportunities to address simultaneously the laser induced quenching of magnetic order for "localized" 4f and for "itinerant" 3d spins. Furthermore, their magnetic properties, such as the magnetic anisotropy, their temperatures of magnetic order, their magnetization, are controllably tunable with the concentration and the layer thickness. It is then possible to monitor the modification of the induced ultrafast dynamics of TM3d and RE4f spins in RE-TM alloys upon the controlled variation of these magnetic properties in order to challenge the predictions of theoretical models [KOO10, BAT10].

The investigations of RE 4f and TM 3d spin dynamics in RE-TM alloys require element- and time-resolved experiments. In my presentation, I will give an overview of our recent results obtained by mean of time-resolved X-ray Magnetic Circular Dichroism (TR-XMCD) experiments performed at large scale facilities either on the femtoslicing beamline at synchrotron BESSYII [BER14, FER17, FER19] or on the TEMPO beamline at synchrotron SOLEIL [SIL17].

[BEA96] Beaurepaire et al. Phys. Rev. Lett 76, 4250, (1996)
[KOO10] Koopmans et al. Nature Mater. 9, 259 (2010)
[BAT10] Battiato et al. Phys. Rev. Lett 105, 027203 (2010)
[BER14] Bergeard et al. Nature Communications 5, 3466 (2014)
[FER17] Ferté et al. Phys. Rev. B 96, 134303 (2017) and Ferté et al. Phys. Rev. B 96, 144427 (2017)
[FER19] Ferté et al. Jour. Magn. And Magn. Mat. 485, 320 (2019)
[SIL17] Silly et al. J. Synchrotron Rad. 24, 886 (2017)

Les moments angulaires de spin et orbital de la lumière

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Une signature première de la lumière est sa fréquence, qui fixe le quanta d'énergie et par là même le quanta d'impulsion d'un champ lumineux. On présentera différents aspects d'une autre propriété fondamentale de la lumière, son moment angulaire, qui peut être de différentes natures: spin ou orbital. La première contribution est associée à l'état de polarisation alors que la deuxième est associé aux degrés de liberté d'espace. A travers quelques exemples, nous verrons ce qui les unit et ce qui les distinguent, et pourquoi l'interaction lumière-matière en présence de moment angulaire attire un intérêt croissant depuis maintenant une trentaine d'année.