Quantum Excitronics : Chiral Emission and Excitation of Single Excitons in TMD-Type Materials

PhD supervisor(s)	Aurelien CUCHE, CNRS Researcher – Gonzague AGEZ, UT3 Researcher e-mail: aurelien.cuche@cemes.fr - agez@cemes.fr group: NeO team of CEMES at Toulouse.
Location	CEMES 29 rue Jeanne Marvig, 31000 TOULOUSE – FRANCE

Summary : This PhD project aims to revolutionize the control, the emission and the two-dimensional diffusion of excitons in 2D materials within the quantum regime. By using first : single-photon sources based on nanodiamonds as a mean to excite single excitons, and second : Bragg–Berry chiral mirrors to demonstrate spin symmetry breaking mechanisms leading to highly circularly polarized light emission, this PhD aims to pave the way for integrated and reconfigurable quantum excitronic devices for quantum computing and communication.

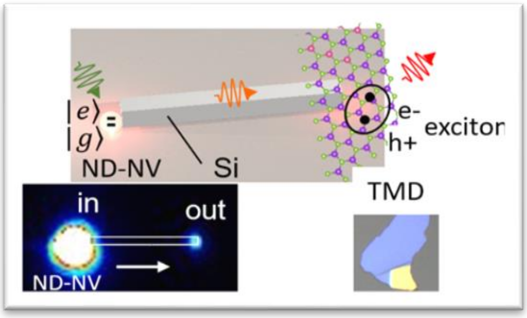
Introduction and context - In order to face the future challenges in communication and data processing, integrated optics and in particular nanophotonics offers deep integration at the nanoscale and is an appealing approach that might lead to disruptive technologies. In this context, Transition Metal Dichalcogenide (TMD) monolayer [1,2] have gained significant attention due to their unique opto-electronic properties. These materials exhibit stable excitons (a neutral electron-hole particle) that diffuse in the material and can recombine radiatively in the far field [3].

Both the phenomena of single exciton diffusion in 2D materials, and their polarization-locked radiative recombination, both from a fundamental and applied point of view, are crucial for applications in quantum technologies. Indeed, understanding and controlling exciton propagation enables the optimization of material and nanostructure design, while pushing the boundaries of nanoscale integration. On the other hand, circularly polarized light emission is a key resource for emerging technologies such as spin-based photonics, quantum communication, and optical information processing.

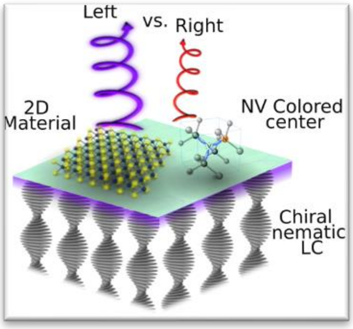
Nevertheless, future exciton-based devices, require precise control over the generation and manipulation of individual excitons, and generation of highly pure circular polarization directly at the exciton source. For a practical implementation, persistent challenges include :

- Precise control of their generation (single-photon optical excitation).
- Manipulation of their quantum state (coherence, spin, valley).
- Control of the polarized far-field emission
- Ultra-sensitive detection of their optical response.

In this PhD project, we propose to develop in parallel two innovative approaches to achieve such a control.



First, by using of single photon emitter colored centers in diamond nanoparticles as an intermediate source for the generation of single excitons [4]. One possible strategy will rely on the use of single-photon sources based on color centers in nanodiamonds (ND-NV), coupled with silicon nanowires [5], to deterministically excite excitons in TMDs. This method decouples photon generation, their propagation, and exciton excitation, paving the way for integrated and reconfigurable silicon-based quantum devices.



Second, we propose a novel paradigm based on near-field coupling between a TMD monolayer and Bragg–Berry chiral mirrors made of cholesteric liquid crystals [6]. These structures exhibit unique properties: they preserve light helicity upon reflection while inducing a spin flip, leading to unconventional chiral optical feedback at the nanoscale. Placing valley-dependent excitons in a TMD in close proximity to such chiral substrates enables a regime where evanescent fields dominate, allowing access to otherwise forbidden optical states and inducing strong asymmetries in polarization-dependent emission dynamics.

This thesis offers therefore an exciting opportunity to engage in cutting-edge research at the intersection of quantum technologies, chiral- and nano-materials, and optoelectronics. By working on the excitation of single excitons in TMD materials using single-photon sources, and on the polarized far-field emission of excitons controlled by Bragg–Berry chiral mirrors, the student will contribute to the development of next-generation quantum devices and gain valuable hands-on experience in experimental physics and nanotechnology.

Thesis Objectives and Methodology - The main objectives of this thesis are :

- **Development of an experimental platform for the generation and study of single excitons -- optimization of the coupling between ND-NVs (single-photon sources) and TMDs via silicon nanowires:** It includes : optical characterization of diamond nanoparticles with colored centers (NV - Nitrogen Vacancy) already coupled to silicon nanowires / Subsequent and controlled deposition of TMD on the same sample. Fabrication of new silicon nanowire samples (via e-beam lithography) and subsequent deposition of TMD monolayer (ExfoLab platform).
- **Development of an experimental hybrid chiral platform :** Development of Bragg–Berry chiral substrates based on cholesteric liquid crystals with tunable photonic bandgaps. Controlled deposition of TMDs using advanced stamping techniques. Integration of h-BN spacers to precisely tune emitter–substrate distance (ExfoLab platform).

- **Optical Characterization :** Analyze of the excitonic species (bright/dark excitons, biexcitons, trions) and their respective dynamics in the TMD. **Single exciton excitation :** Investigation of the excitation mechanisms of single excitons in TMD materials using single photons emitted by the diamond nanoparticles by performing low-temperature (4K - allowing for the suppression of thermal effects and enhanced resolution in the detection of the excitonic state) optical characterizations (photoluminescence, time-resolved measurements, Fourier plane imaging, correlation measurement), to analyze the behavior of single excitons in the TMD materials. **Chiral emission :** Polarization-resolved photoluminescence measurements to quantify circular dichroism and dissymmetry factors. Time-resolved spectroscopy to probe modifications of the local density of optical states (LDOS), at room temperature and 4K cryogenic conditions. Fourier plane imaging to analyze emission directionality and spin-dependent radiation patterns.
- **Analysis of results:** Detailed analysis and comparison of the optical properties of the TMD materials in the classical and quantum regimes. **Single exciton excitation :** The emitted photons and the resulting excitonic behavior will be analyzed using various techniques, including spectroscopy, to understand the efficiency and dynamics of exciton generation. Results will be compared to electrodynamic simulations (FDTD, GDM) in order to optimize the photon-exciton coupling, while single exciton diffusion will be model numerically using homemade simulation tools. **Chiral emission :** Investigation of how the chiral environment modifies exciton recombination dynamics, spin states, and emission helicity. Study of distance-dependent coupling regimes to identify signatures of near-field symmetry breaking. Numerical simulations (FDTD) to model emitter–substrate interactions.
- **Integration and Demonstration :** Fabrication of excitronic prototype devices and functionality tests (quantum switch, optical memory, chiral quantum light source).

Expected Results and Impact - The outcome of this project is expected to provide a fundamental understanding of the mechanisms leading to the possible excitation of excitons in TMD by single photons and their interaction. It will lead to the redaction of publications in international journals. This work will contribute to the development of novel optoelectronics devices for quantum technologies, particularly in the field of quantum information processing.

References: [1] G. Wang et al. Rev. Mod. Phys. 90, 021001 (2018). [2] M Poumirol et al. ACS Photonics 7, 11, 3106 (2020). [3] H. Lamsaadi et al. Advanced Optical Materials, 13 (10), 2403009 (2025). [4] C. Bradac et al. Nat. Comm. 10, 5625 (2019). [5] M. Humbert et al. Phys. Rev. Applied 17, 014008 (2022). [6] G. Agez et al. Optica, 9 (6), 652-655 (2022).

Keywords, areas of expertise	Quantum technologies, single photon, colored centers, nanodiamond, TMD monolayer, exciton, chirality, time-resolved optical microscopy, photon correlation, numerical simulation of optical properties, simulations.
Required skills for the PhD thesis	The student will be involved in the experimental aspects of the optical characterization of optical excitation (down to single photon excitation) and emission of TMDs, and to the theoretical aspects of such light-matter interaction (electrodynamics simulation).

	Education: Master's degree in physics (specialization in quantum optics, nanophotonics, condensed matter materials science) or equivalent. Autonomy, dynamism, scientific curiosity and rigor are the key words to carry out this project. Advanced understanding of optics and materials science, especially related to semiconductors (TMD) materials. Familiarity with laboratory work, programming (Python). Knowledge or experience in optical spectroscopy techniques, especially for low-temperature measurements.