ÉCOLE DOCTORALE GALILÉE
Université Sorbonne Paris Nord

Contrat doctoral – ED Galilée

Titre du sujet : Exciton Physics in Organic Semiconductor Laser materials

Physique des Excitons dans des matériaux semiconducteurs organiques pour lasers.

- Unité de recherche : Laboratoire de Physique des Lasers (LPL), Institut Galilée, Université Sorbonne Paris Nord et CNRS (UMR 7538)
- Discipline : Physique
- Direction de thèse : Sébastien Forget
- Contact : Sébastien Forget
- Domaine de recherche : Photonique
- Mots clés : Lasers, photophysique, matériaux organiques

Résumé court du projet (en français et en anglais) :

La photonique et l'optoélectronique organiques sont des domaines émergents ayant conduit à plusieurs avancées récentes dans les domaines du photovoltaïque, de l'éclairage ou des displays. Les lasers restent encore à l'état de développement embryonnaires. La clé des progrès réside dans la disponibilité de matériaux efficaces qui seraient idéalement spécifiquement "conçus" pour répondre aux exigences du fonctionnement en régime laser. Mais un bon matériau luminescent (pour les OLED par exemple) n'est pas nécessairement un bon matériau pour le laser, et vice versa. Jusqu'à présent, la compréhension fondamentale de ce qui fait un "bon" matériau organique pour les lasers fait toujours défaut. Pour aller plus loin dans cette direction cruciale, une collaboration forte entre chimistes organiques, physiciens et technologues est nécessaire.
Ce projet de thèse est <u>pluridisciplinaire</u> car il fait travailler ensemble opticiens (au LPL), chimistes (synthèse de molécules organiques à l'IPCM), et technologues à l'Université de Kyushu au Japon. Il est <u>international</u> car trois séjours de longue durée au Japon sont prévus, financés par un IRP et par le partenaire japonais.

Organic photonics and optoelectronics are emerging fields that have led to several recent advances in areas such as photovoltaics, lighting, and displays. However, lasers are still in the early stages of development. The key to progress lies in the availability of efficient materials that would ideally be specifically "designed" to meet the requirements for laser operation. However, a good luminescent material (for example, for OLEDs) is not necessarily a good material for lasers, and vice versa. Until now, there has been a fundamental lack of understanding of what makes a "good" organic material for lasers. To further progress in this crucial direction, strong collaboration between organic chemists, physicists, and technologists is necessary.
This thesis project is multidisciplinary as it involves collaboration between optics researchers (at LPL), chemists (organic molecule synthesis at IPCM), and technologists at Kyushu University in Japan. It is international in scope as it involves three long-term stays in Japan, funded by an IRP and the Japanese partner.

Sujet complet (en anglais) :

Organic optoelectronics and photonics is an emerging domain that led to several recent breakthroughs in the fields of photovoltaics, solid-state lighting and displays. Organic Light-Emitting Diodes (OLEDs), based on luminescence of organic semiconductors, are already present in most commercial smartphone displays owing to their remarkable 100% internal quantum efficiencies. On the other hand, **organic laser diodes** (relying on stimulated emission instead of spontaneous emission), have just been demonstrated, at a burgeoning stage, under direct electrical pumping, which makes it a much more exploratory topic. They represent both a fundamental challenge to overcome and a potential source of new applications for instance in bio-sensing. Unlike traditional inorganic semiconductors whose emission wavelengths are fixed to a small set of available fixed energy bandgaps, organic semiconductors are excitonic materials that can be easily tuned by chemical design to cover the whole visible spectrum. Furthermore, while traditional diode lasers are based on epitaxial techniques, organic diode lasers can be made from solution or low-temperature evaporation techniques. This enables a fabrication process at lower energy costs and with limited usage of non-renewable resources, and guarantees an easy pairing with all existing technological platforms.

Very recently, **important breakthroughs have been reported in the field**, in particular progresses towards electrical driving[1,2]. In parallel, it was also shown that inserting a light-emitting organic semiconductor inside an optical cavity is an effective way to create strong coupling of photons and excitons, showing another way to achieve lasing through cavity polaritons[3] or polariton condensates instead of simply photons, with the promise of lower thresholds.

In these examples, the key for progress relies in the availability of efficient materials that would be ideally specifically "designed" to meet the requirements of laser operation. However, a good luminescent material (for OLEDs for instance) is not necessarily a good material for lasing, and vice-versa. **Up to now the fundamental understanding of what makes a "good" organic material for lasers is still lacking**. To go a step further in this crucial direction, a strong collaboration between organic chemists and physicists is required.

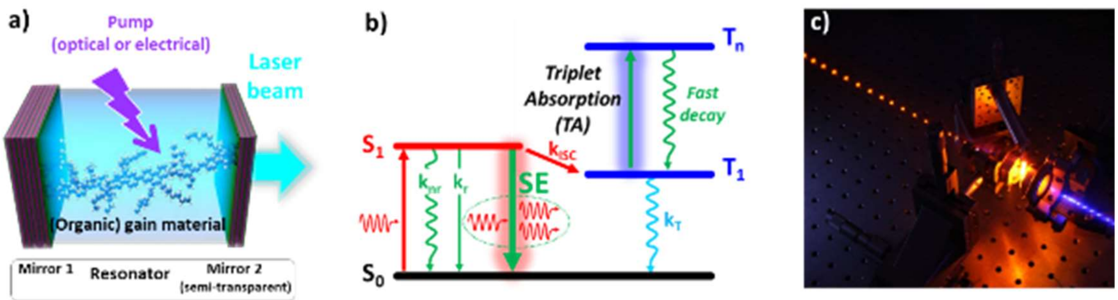

Figure 1: a) General structure of a laser. b) Simplified Jablonski diagram with dominant mechanisms under optical pumping: SE = stimulated emission, k_r/k_{nr} = radiative/nonradiative decay rates of singlet state, k_T = triplet state decay rate, k_{ISC} = intersystem crossing rate. c) Image of optically-pumped organic Laser beam (LPL).

<u>Context of the thesis</u>

This PhD is proposed within the framework of an **international project** aiming at developing organic lasers and organic semiconductor-based quantum devices. The project gathers chemists, physicists and nanofabrication specialists, within a fruitful collaboration connecting laser physicists at LPL, photo physicists in Strasbourg (IPCMS), and chemists at IPCM in Paris. Alongside, an International Research Project (IRP funded by CNRS) between the French partners and a world-leading laboratory in Japan (the Adachi group at Kyushu university), enables a full access to state-of-the-art facilities, materials, and characterization tools.

In this project, our chemist partners will synthetize various polymers/small molecules with presumably good properties for lasing. In our group of physicists at LPL, we will design and build experiments in order to understand and measure the adequate optical properties of those materials, enabling a fruitful feedback and enhanced understanding of the link between molecular structure and lasing properties.

During the PhD first year, the candidate will build a new setup for characterizing gain materials — based on the measurement of Amplified Spontaneous Emission (ASE) threshold — in order to make it more reliable and to measure some photophysical data associated to triplet excitons, such as triplet absorption cross sections or singlet-triplet exciton annihilation rates. These phenomena are among the major obstacles hindering organic laser diodes, and can theoretically be observed and measured from a single high-intensity, long-pulse, stripe-shaped diode excitation, but this has never been observed yet.

During the second and third year, the objective will be to build a novel instrumentation platform combining several experimental techniques to study the exciton dynamics in organic semiconductors for lasing, including the previously mentioned ASE experiment, but also new pump-probe and time-resolved experiments for triplet absorption and triplet state lifetime measurements. The objective will be twofold: 1) scanning organic materials to select the better ones for lasing in a comprehensive way 2) using those materials for the fabrication (potentially in clean room environment) and characterization of very efficient lasing devices. During the PhD, **several research stays in Japan will be necessary**.

Applicant skills: A strong background in optics and general physics, and notions in lasers are needed. No specific skills in organic electronics or chemistry are required, but the candidate needs being open-minded and curious as numerous interactions with our chemist partners are foreseen. Given the pluridisciplinarity and international character of the project, the candidate (during the PhD) will play a central role between partners and consequently good communication skills are required. The work is mostly experimental but does not consist in a single “big” experiment but rather on several more simple setups. Therefore, it is expected that the candidate participates in all aspects of research, including (at the PhD level) simulation work and paper writing.

References :

[1] A.S.D. Sandanayaka, T. Matsushima, F. Bencheikh, S. Terakawa, W.J. Potscavage, C. Qin, T. Fujihara, K. Goushi, J.-C. Ribierre, and C. Adachi, Appl. Phys. Express **12**, 061010 (2019).

[2] K. Yoshida, J. Gong, A.L. Kanibolotsky, P.J. Skabara, G.A. Turnbull, and I.D. W Samuel, | Nature | **621**, (2023).

[3] T. Ishii, F. Bencheikh, S. Forget, S. Chénais, B. Heinrich, D. Kreher, L. Sosa Vargas, K. Miyata, K. Onda, T. Fujihara, S. Kéna-Cohen, F. Mathevet, and C. Adachi, Adv. Opt. Mater. **9**, 1 (2021).