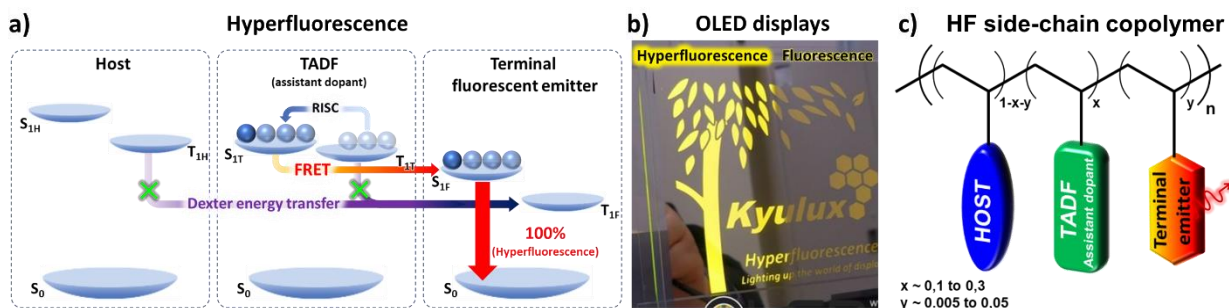


## Self-organized hyperfluorescent materials for next generation of optoelectronic devices

**KEYWORDS:**  $\pi$ -conjugated polymers, liquid crystals, hyperfluorescence, OLEDs, optoelectronics.

**1. CONTEXT & PROJECT GOALS:** Hyperfluorescence (HF) for OLEDs, first proposed by the group of Prof. Chihaya Adachi,<sup>[1]</sup> offers a promising route to realize high-efficiency electroluminescence EL with better operational stability and excellent colour purity (Figure 1a and 1b). HF organic light emitting diodes (OLEDs) use thermally activated delayed fluorescence (TADF) molecules as assistant dopants and combine the high efficiency and triplet harvesting ability of these TADF donors<sup>[2]</sup> with the stability and colour purity of conventional fluorescent acceptor molecules (Figure 1a). The host, TADF and fluorescent molecules, which are usually blended by thermal co-evaporation to form the HF emitting layers (EMLs), must be carefully selected to allow effective Förster resonance energy transfer (FRET) of all electrically generated singlet and triplet excitons from the TADF assistant dopants to the singlet excitons of the fluorescent emitters. Such an approach has demonstrated numerous OLEDs with internal quantum efficiency (IQE) approaching 100% and improved operational stability compared to purely TADF devices.<sup>[3]</sup> The precise control of the intermolecular interactions between the three components of the HF-EMLs is crucial for achieving excellent EL performance. In particular, the suppression of Dexter energy transfer from the triplet state of the TADF assistant dopant to the non-emissive triplet state of the fluorescent emitter is essential to reduce energy losses and realize HF OLEDs with high IQE, low efficiency roll-off, and extended device lifetime. To date, hyperfluorescent systems for OLEDs have been almost exclusively based on molecular systems, with only rare examples of hyperfluorescent macromolecular systems reported.<sup>[4]</sup> Furthermore, only amorphous systems have been developed, leading to isotropic emitting system and consequently significant light losses in emitting devices.



**Figure 1.** a) Emission mechanism of TADF and schematic representation of the energy transfer mechanism of hyperfluorescence. b) Comparison of hyperfluorescent and fluorescent OLEDs based on the same yellow emitter. c) Design of hyperfluorescent copolymers incorporating in a same backbone different ratio of host (matrix), TADF dopant, and terminal emitter monomers.

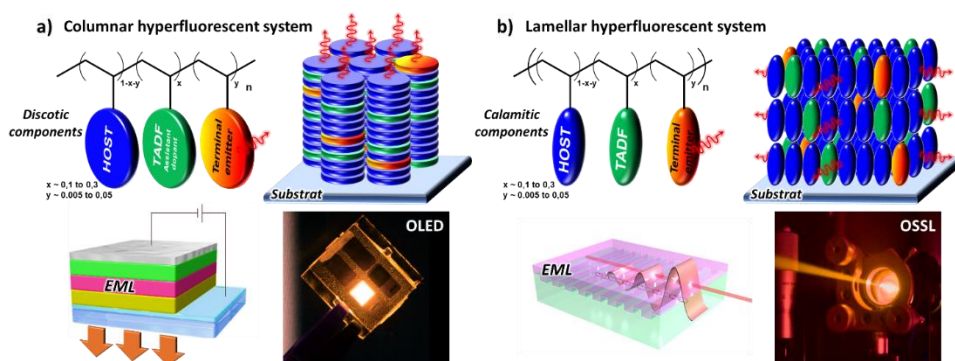
In this context, we propose in this international project to **design and investigated novel self-organized hyperfluorescent materials based on liquid crystalline (LC) macromolecular systems for light emitting device applications such as OLEDs and organic solid-state lasers (OSSLs).** It will be developed via the combination of expertise in material design & synthesis (IPCM, SU), soft matter and thin film characterisation (INSP, SU) **in France**; and the world-leading group in TADF and hyperfluorescent OLEDs (OPERA, Kyushu University) **in Japan**.

## 2. JUSTIFICATION OF SCIENTIFIC APPROACH:

This research project offers a novel and more-efficient approach to design anisotropic hyperfluorescent systems in which, the orientation of the emitting components in the EMLs of the optoelectronic devices will be controlled via the mesomorphic properties of the target emitting copolymers.

### Material Synthesis (experts: L. Sosa Vargas, F. Mathevet, SU):

These LC hyperfluorescent materials will be based on discotic (disk-like) or calamitic (rod-like) host, TADF dopant and terminal fluorescent monomers dispersed in different ratios along a polymer backbone to form columnar or lamellar macromolecular architectures (Figure 2). In such anisotropic systems, the light emission will be fully oriented perpendicularly or parallel to the plane of the films, leading to highly emissive EMLs for



**Figure 2.** Schematic representation of a) columnar and b) lamellar hyperfluorescent macromolecular systems, based on discotic or calamitic components, respectively, with preferential light emission perpendicular or parallel to the EML for OLED or OSSL applications.

OLED and OSSL devices. Taking advantage of the expertise in the group,<sup>[5]</sup> the PhD candidate will be responsible for designing and synthesizing the different mesogenic monomers, synthesizing the target copolymers, and conducting both their preliminary LC and photophysical characterizations at IPCM (SU).

**Characterisation of thin film and bulk properties (expert: E. Lacaze, SU):** Advanced structural and morphological characterisations of the thin films of the materials will be performed by atomic force microscopy (AFM), X-ray diffraction XRD and synchrotron-based grazing incident wide-angle X-ray scattering (GIWAXS) in collaboration with INSP (SU). The PhD candidate will carry out these characterizations in close collaboration with Dr. E. Lacaze, an expert in LC thin film investigations, who will be deeply involved in this project.<sup>[6]</sup>

**Photophysical characterization and device fabrication (expert: C. Adachi, Kyushu U):** The advanced photophysical characterizations of the new LC hyperfluorescent copolymers in the solid state (photoluminescence quantum yield (PLQY), fluorescence lifetimes, ...) will be carried out by the PhD candidate in Japan through several long stays at the OPERA center (2x6 months) under the supervision of Prof. Chihaya Adachi (Japanese co-supervisor). Specific measurements, such as time-resolved spectroscopy as a function of temperature in the solid state, as well as angle-dependence PL measurement, will also be performed at the Japanese partner laboratory. Ultimately, the PhD candidate will fabricate and optimize OLEDs and OSSLs to evaluate the electroluminescence and laser properties of these new materials (Figure 2), leveraging OPERA's state-of-the-art facilities and the unique expertise of Prof. Adachi's group in this field.<sup>[1,2,3,9]</sup>

**PARTENARIAT STRATEGIQUE and MANAGMENT of the Project:** For almost ten years, a strong and dynamic collaboration has been established between the OPERA center and French researchers from IPCM (SU) in the field of organic optoelectronics. These interactions have taken place within the framework of collaborative research projects, led by both Japanese and French partners (ERATO-JST program 2014-2019, JSPS Core-to-Core program 2018-2023, JSPS ILR project 2023-2030, CNRS-PICS 2019-2021). Since 2015, these long-standing and expanding collaborations between OPERA and IPCM have led to an increasing number of joint research efforts, resulting in 27 co-authored publications and 2 CNRS-SU-KU joint patents to date. Based on these excellent results, an International Research Project (IRP) with OPERA was launched by the CNRS in January 2023 for a duration of five years. This IRP, named *LUX-ERIT*, focuses on the development and integration of new organic semiconductors and light emitters for optoelectronics and photonics. It brings together four French laboratories (IPCM-SU, ILV-USVQ, IPCMS-UDS, and LPL-USPN), with Dr. F. Mathevet as the French coordinator and Professor Chihaya Adachi as the Japanese coordinator. This IRP has facilitated numerous exchanges, including visits by researchers, postdocs, and students from the different French partners to the OPERA center, fostering active knowledge transfer and joint scientific advancements. These efforts have already led to the publication of 10 joint articles and the submission of a patent related to organic lasers. Notably, a proposal for the creation of an International Research Laboratory (IRL) with the OPERA center, involving several IPCM members, was submitted to CNRS in February 2025 for a potential launch in January 2026, pending approval. In addition to the strong scientific expertise of the French and Japanese supervisors of this project and their extensive experience in collaboration, the PhD candidate will benefit from this dynamic research environment, fostering close interactions and knowledge exchange between the two partners.

## REFERENCES

- [1] H. Nakanotani, T. Higuchi, T. Furukawa, K. Masui, K. Morimoto, M. Numata, H. Tanaka, Y. Sagara, T. Yasuda, C. Adachi, *Nat. Commun.*, **2014**, 5, 4016.
- [2] H. Uoyama, K. Goushi, K. Shizu, H. Nomura, C. Adachi, *Nature*, **2012**, 492, 234.
- [3] Y.-T. Lee, C.-Y. Chan, N. Matsuno, S. Uemura, S. Oda, M. Kondo, R. W. Weerasinghe, Y. Hu, G. N. Iswara Lestanto, Y. Tsuchiya, Y. Li, T. Hatakeyama, C. Adachi. *Nat. Commun.*, **2024**, 15, 3174.
- [4] J. Hu, Y. Wang, Q. Li, S. Shao, L. Wang, X. Jing, F. Wang. *Chem. Sci.*, **2021**, 12, 13083.
- [5] a) L. Chen, C. Li, E. Fu, M. Li, Y. Kuboi, Z.-Y. Li, Z. Chen, J. Chen, X. Liu, X. Tang, L. Frederic, F. Maurel, C. Adachi, F. Mathevet, S. Zhang. *ACS Materials Lett.*, **2023**, 5, 1450. b) M. Auffray, D. H. Kim, J. U. Kim, F. Bencheikh, D. Kreher, Q. Zhang, A. D'Aléo, J.-C. Ribierre, F. Mathevet, C. Adachi. *Chem. Asian J.*, **2019**, 14, 1921. c) 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, C. Adachi. *Adv. Opt. Mater.*, **2021**, 9, 2101048. d) Y. Xiao, D. Zeng, L. M. Mazur, A. Castiglione, E. Lacaze, B. Heinrich, B. Donnio, D. Kreher, A.-J. Attias, J.-C. Ribierre, F. Mathevet. *Polymer Journal*, **2017**, 49, 31.
- [6] a) J. D. Niyonzima, H. Jeridi, L. Essaoui, C. Tosarelli, A. Vlad, A. Coati, S. Royer, I. Trimaille, M. Goldmann, B. Gallas , D. Constantin, D. Babonneau, Y. Garreau, B. Croset, S. Kralj, R. D. Kamien, E. Lacaze, *Phys. Rev. Lett.* **2025**, 134, 018101. b) I. Gharbi, V. Palacio-Betancur, H. Ayeb, D. Demaille, J. J. de Pablo, R. D. Kamien, E. Lacaze. *ACS Appl. Nano Mater.* **2021**, 4, 7, 6700. c) B. Rožič, J. Fresnais, C.eline Molinaro, J. Calixte, S. Umadevi, S. Lau-Truong, N. Felidj, T. Kraus, F. Charra, V. Dupuis, T. Hegmann, C. Fiorini-Debuisschert, B. Gallas, E. Lacaze. *ACS Nano*, **2017**, 11, 7, 6728.
- [9] a) C.-Y. Chan, M. Tanaka, Y.-T. Lee, Y.-W. Wong, H. Nakanotani, T. Hatakeyama, C. Adachi. *Nature Photonics* **2021**, 15, 203. b) T. Matsushima, F. Bencheikh, T. Komino, M. R. Leyden, A. S. D. Sandanayaka, C. Qin, C. Adachi. *Nature* **2019**, 572, 502. c) C. Qin, A. S. D. Sandanayaka, C. Zhao, T. Matsushima, D. Zhang, T. Fujihara, C. Adachi. *Nature* **2020**, 585, 53.