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Financement : Société Française de Microscopie

Experimental development of photoluminescence and Raman Spectroscopies within an Electron Microscope for in-situ studies of electron induced transformation of dye molecules

Luminescence in a TEM as single molecule fingerprint: The luminescence signature of molecules is sensitive to its electronic configuration, skeletal orientation (*e.g.*, isomers and rotamers), internal relaxation processes (*e.g.*, vibrations), neighboring interactions (*e.g.*, energy and electron transfer processes), and is established at sensitivities down to the single-molecule level^{1,2}. In the same fashion Raman spectroscopy is used routinely for the tracking of the vibrational fingerprint.

Thus, the ability to simultaneously measure both the photoluminescence (PL) and vibrational fingerprints of molecules within an electron microscope would enable real-time monitoring of electron-molecule interactions, offering a comprehensive understanding of fundamental electron-induced molecular physics and chemical transformations.

The goal of this internship is to build an in-situ photoluminescence (PL) and Raman set-up on a HF2000 (scanning) transmission electron microscope ((S)TEM) equipped with a parabolic mirror for injection and collection of light³, as used routinely for cathodoluminescence⁴ (**Figure 1**). With this setup, first proof-of-principle experiments will be performed on dye molecules.

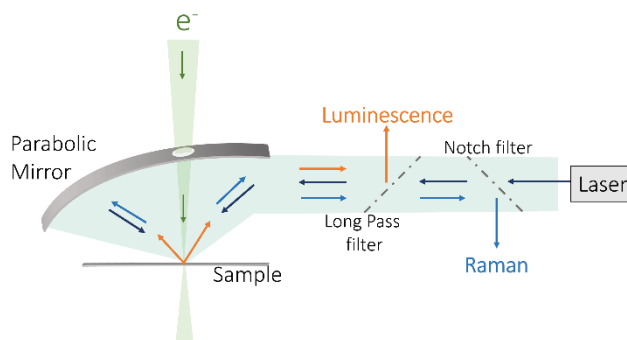


Figure 1: Light injection and collection set-up in a (S)TEM equipped with a parabolic mirror.

In this project, located at the interface between Physics and Chemistry, the student will learn:

- 1) **Instrumentation** (building of a custom-made light path for light collection compatible with light injection, Figure 1).
- 2) **Raman and Photoluminescence spectroscopy** (from the building of a setup to the interpretation of the results).
- 3) **Fundamental understanding of electron-matter interaction** (and the radiation transformation through an original technique of in-situ light spectroscopy in electron microscopy).
- 4) **Electronic excitation and decay pathways** (in molecules upon e-beam irradiation).

1. Kondo, T., et al. Single-Molecule Fluorescence Spectroscopy of Photosynthetic Systems. *Chem. Rev.* **117**, 860–898 (2017).
2. Dos Santos, J. M. et al. The Golden Age of Thermally Activated Delayed Fluorescence Materials: Design and Exploitation. *Chem. Rev.* **124**, 13736–14110 (2024).
3. Tizei, L. H. G., et al, M. Cathodoluminescence, light injection and EELS in STEM: from comparative to coincidence experiments. *Microscopy* **75**, 129–144 (2026).
4. Coenen, T. & Haegel, Cathodoluminescence for the 21st century: Learning more from light Appl. Phys. Rev. **4**, 031103 (2017)