Unraveling Hot Carrier Processes in Plasmonic Energy Devices

Giulia Tagliabue[1]

1. Laboratory of Nanoscience for Energy Technologies (LNET), School of Engineering, Ecole Polytechnique Federale de Lausanne (EPFL)

E-mail: giulia.tagliabue@epfl.ch

In the last decade, plasmonic and dielectric nanoantennas have revolutionized light manipulation and control at the nanoscale. Interestingly, hot carriers and photoluminescence in metals have opened new pathways for controlling photo(electro)chemical processes and monitoring temperatures. Yet, fundamental questions remain about the microscopic details of these complex light-matter interactions.

We have recently developed a well-controlled experimental platform based on ultrathin monocrystalline gold flakes [1] that, in combination with theory, enables deeper insight into light absorption and emission processes. In particular, I will show results unraveling the origin of luminescence in gold, exploring the role of physical confinement, excitation wavelength and temperature [2] and I will also show how crystallinity improves hot carrier dynamics and transport [3]. I will then present micro-scale photoelectrochemical measurements clarifying the interplay of hot carrier generation/transport and quantifying the injection probability of high-energy d-band holes at the metal-electrolyte interface [4], connecting it to the ultrafast dynamics of these carriers in monocrystalline metals. Finally, I will briefly discuss light-driven thermo-optical effects in dielectric nanoantennas and their potential to control temperature fields at the nanoscale [5]. Overall, this microscopic insight is critical to advance the design of plasmonic-based energy devices.

References

[1] Kiani F., Tagliabue G. - High-Aspect Ratio Au Microflakes via Gap-Assisted Synthesis – Chemistry of Materials 34 (3), 1278-1288

[2] Bowman A.R., Echarri A. R., Kiani F., Iyikanat F., Tsoulos T.V., Cox J. D., Sundararaman R., Garcia de Abajo J., Tagliabue G. - Quantum-mechanical effects in photoluminescence from thin crystalline gold films – arXiv 2307.08477

[3] CO Karaman, AY Bykov, F Kiani, G Tagliabue, AV Zayats - Ultrafast hot-carrier dynamics in ultrathin monocrystalline gold - Nature Communications 15 (1), 703

[4] Kiani F, Bowman A.R., Sabzehparvar M., Karaman C.O., Sundararaman R., Tagliabue G. - Transport and Interfacial Injection of d-Band Hot Holes Control Plasmonic Chemistry – ACS Energy Letter, 2023, 8, 4242–4250

[5] Naef A., Mohammadi E., Tsoulos T., Tagliabue G. - Light-Driven Thermo-Optical Effects in Nanoresonator Arrays – Advanced Optical Materials 2023