

PhD thesis project: Modelling non-linear photoluminescence in 2D plasmonic cavities

Supervisor(s): Gérard COLAS des FRANCS (ICB, Dijon France)
Yonatan SIVAN (Ben Gurion University, Israël)

Laboratory: ICB

Metal nanostructures support so-called surface plasmons polaritons (SP), coupling collective oscillations of the free electrons of the metal (plasmon) to an electromagnetic wave (polariton). SP enables strong subwavelength confinement of energy, which is of great interest for compact on-chip all-optical devices.

We are developing SP-based ALUs (arithmetic and logic units) that perform chosen complex Boolean functions (AND, OR, XOR, etc. logic gates). These are smaller and potentially largely faster than electronics microprocessors. Recently, we have implemented an all 2-bit logic gates and a 1-bit full adder on a single reconfigurable plasmonic structure [1,2] and are currently working on finding optimal configurations. The Boolean information inputs are carried by the polarization of an infra-red pulse which are processed by the SP modes towards the output ports where they are converted into Boolean output signals encoded in the non-linear photoluminescence (NPL) response. To model this complex process, we performed numerical simulations of the linear near-field plasmonic transmittance response and a phenomenological model for conversion from near-field to NPL maps (see Fig. 3 and Supp. Info of ref. [2]). Although this permits a qualitative interpretation of the measured NPL images, a more quantitative model is needed to accurately compute the non-linear transmittance of the plasmonics ALUs. Moreover, this will help to develop with our partner (CIAD lab, Dijon) a composite artificial intelligence, which combines physics-informed machine learning with domain ontologies and evolutionary strategies. This will allow a realistic optimization procedure and expanding our approach to more complex all-optical computing units.

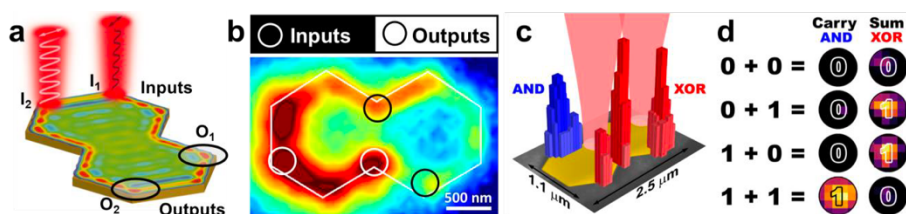


Figure 1. (a) Scheme of a Double Hexagon (DH) plasmonic ALU with 2 inputs (I_1 , I_2) and 2 outputs (O_1 , O_2). The SP is watermarked on the structure. (b) Experimental ('0' - '1') optical NPL response of a gold DH device. (c,d) Experimental output of the AND and XOR signals realized simultaneously. (d) The dual read-out for the four excitation configurations realizes the full 1+1 bit adder in the same DH.

Photoluminescence from noble metals is a well-known phenomenon but its generation mechanism, especially pulsed excitation, has been debated [3,4]. Significant progress has been obtained recently in the theoretical understanding of light emission from metals, mainly for *uniform* excitation and in *the continuous regime* [5-9]. Specifically, a formula was established for the emission probability of an excited electron in the metal. The formula requires only knowledge of the local electric field and the electron temperature. Heuristic extension of NPL modelling from continuous to pulsed laser is generally done considering a quasi-stationary approximation with a time dependent electron

distribution. To predict more accurately the non-linear light emission from the ALU cavities and to extend it to scenarios of non-uniform excitation of the plasmonic system (see Fig. 1(a)), we will extend the analytic approach from the steady-state (CW illumination) to the transient case (pulsed illumination) by tracking the dynamics of the electron distribution, the field and electron temperature for each excitation pulse intensity and duration. We will also combine the knowledge on the transport properties of the non-thermal electrons to account for the delocalized nature of the emission observed experimentally.

References

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- [2] *Compact implementation of a 1-Bit Adder by Coherent 2-Beam Excitation of a single Plasmonic Cavity*, Dell'Ova *et al*, ACS Photonics **11**, 752 (2024)
- [3] *Dynamics, Efficiency, and Energy Distribution of Nonlinear Plasmon-Assisted Generation of Hot Carriers*. Demichel *et al*, ACS Photonics **3**, 791–795 (2016)
- [4] *Spatial Distribution of the Nonlinear Photoluminescence in Au Nanowires*, Agreda *et al*, ACS Photonics **6**, 1240 (2019)
- [5] *“Hot” electrons in metallic nanostructures – non-thermal carriers or heating ?*
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- [6] *Theory of “Hot” Photoluminescence from Drude Metals*,
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- [7] *Nonlinear Photoluminescence in Gold Thin Films*
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- [8] *Theory of photoluminescence by metallic structures*
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- [9] *Photoluminescence from Metal Nanostructures: Dependence on Size*
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Type of project (theory / experiment): theoretical

Required knowledge: Nanophotonics

Funding : ANR (obtained), 2200 €/month (gross salary)

Expected starting date : September or October 2026

Application : Please send **before May 31** to Prof. Gérard Colas des Francs (gcolas@ube.fr) : a CV, a letter of motivation, a contact reference and bachelor/master transcript