



PhD Thesis Defense FRANCISCO PELAYO GARCIA DE ARQUER 'Plasmonic Hot-Carrier Optoelectronics'

FRANCISCO PELAYO GARCIA DE ARQUER

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Wednesday, February 18, 11.00. ICFO Auditorium

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Solution processed nano-photonic devices

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The detection of light is of central importance in both fundamental science and applied

technology. Photodetectors, which aim at transducing optical stimulus into measurable electrical signals, are ubiquitous to modern society, and widespread from home electronics to more sophisticated applications. Harnessing solar light-energy has the potential to replace current environmentally unfriendly fossil fuels, which is of paramount importance for a sustainable development.

Fostered by the strong interaction of light with metal nanostructures, Plasmonics have seen tremendous advances during the last decades. Unique phenomena such as intense electric field enhancement and localization, plus tunable and high absorption across the visible-infrared region of the spectrum are especially attractive for optoelectronic applications. In that framework, plasmonics have been introduced to improve the performance of photodetectors and solar cells by modulating the absorption in the active semiconductor. Another approach consists instead in harnessing the energetic, hot electrons that arise after plasmon excitation in the metal. Within this scheme, unlike in semiconductors, light can be absorbed in the tens of nanometer scale and the optoelectronic spectral response tailored by metal-nanostructuring.

Plasmonic hot-electron optoelectronics has seen a very vivid research during the last years. Early progress focused in the field of photocatalysis, where metal nanoparticles were used to extend to the visible the spectral response of high-bandgap semiconductors. Prior to this thesis, no solid-state plasmonic solar cells had been reported. Further sensitization into the infrared was achieved by employing arrays of metallic nano-antennas in a metal-semiconductor Schottky architecture. The fabrication of these devices relied however on small-area and low-throughput lithographies, which complicates the deployment of this technology for photovoltaic and photocatalytic applications. Moreover, their performance remains yet on the low side.

The aim of this thesis is to further contribute to the development of this novel class of devices, with emphasis in photovoltaic and photodetection applications. We start by identifying the crucial role of the interface on the photovoltaic performance, where we find that surface states hinder energy collection. We show that, by introducing an ultrathin insulating barrier, these defects can be passivated allowing for solar energy harnessing. This comes however at the cost of reduced hot-electron injection. The first part of this thesis is

focused on the interface optimization by both inorganic (chapter 3) and organic (chapter 4) approaches. Inorganic passivation schemes allow for high open-circuit voltages and fill-factors, but the mediocre short-circuit current limits device performance. Organic passivation is achieved by the use of self-assembled monolayers.

(SAMs). By controlling molecule's shape and functionalization both open-circuit voltage and short-circuit current can be tailored, enabling for higher performances and quantum efficiencies that go up to 5%.

In chapter 5 we present a plasmonic crystal architecture to tailor the spectral response of hot-electron plasmonic photodetectors across the visible-near infrared. We identify and exploit the interplay between the different localized and lattice modes in the structure, with responsivities up to 70 mA/W. Notably, this architecture is fabricated by soft-nanoimprint lithography, a low-cost and high-throughput technique, compatible with large-scale manufacturing processes such as roll-to-roll.

We conclude by analyzing the upper bounds in the performance of plasmonic hot-carrier optoelectronic devices based on a metal-insulator-semiconductor architecture and presenting future challenges in the field (chapter 6)

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