

AAP 2022 iMAT - Scientific application form

2D/0D Heterostructure for IR Light Absorption/Detection

1. Scientific context of project

- **Aim, Context, Scientific approach, Risks,** (Bibliography on page 3).

Aim: the project targets to design a mixed dimensionalities van der Waals heterostructure made of a 2D material used as high mobility material and 0D nanocrystals (NCs) used as the optically absorbing material. A first target is to demonstrate light sensitization of the 2D material for energy below its band gap. In such structure the coupling between the two materials is a tunnel coupling. As a result, only the first few layers of NCs in the direct vicinity of the 2D film are transferring charge to the 2D material. More generally, 2D materials present overall low absorption due to their thin character. A second target of this project is to tackle this issue through the introduction of a light resonator which will enhance the light absorption at the absorption wavelength of the NCs. The project will be a collaboration between IMPMC and INSP and will be led by Johan Biscaras.

Context: Graphene has renewed the interest for 2D materials which rapidly expanded to other 2D layers especially transition metal dichalcogenides. The various band gap and conductive character of the 2D materials enable to use them as electrodes, semiconductors and insulators. Among many striking properties, 2D materials are easily functionalized, and thus their coupling to atoms, molecules and nanoparticles have quickly been explored. In 2012, Konstantatos et alⁱ, explored a hybrid structure made of graphene coupled to PbS quantum dots, as represented in the schematic of the device in **Figure 1**. By doing so the Spanish team has demonstrated infrared sensitization of graphene in the near IR. A very striking feature is the very large photoconductive gain of such hybrid structures. Under light illumination, light absorption occurs mostly in the NC layer, band alignment with graphene favors the transfer of one carrier selectively. This charge will recirculate as long as the other charge trapped in the NC remains un-recombined. Thus, large gain is generated and this leads to high responsivity (*ie* ability to convert light into current). Though conceptually new, this hybrid structure suffers from many limitations.

In particular the lack of band gap in graphene leads to a larger dark conductance, which limits the signal to noise ratio. Later it has been proposed to replace the graphene by MoS₂ to solve this dark current limitationⁱⁱ. It has also been proposed to expand the spectral range of this hybrid device by using even narrower band gap NC such as HgTe. To date longest wavelength reported using this approach is 3 μm.ⁱⁱⁱ Now we aim to expand this spectral range toward the 3-5 μm range which is important for applications since it corresponds to an atmospheric transparency window. Thus, in this range long distance imaging and detection become possible.

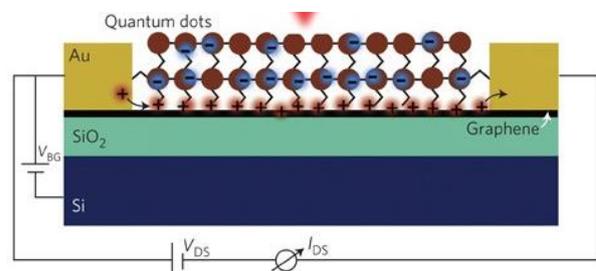


Figure 1 Schematic of a phototransistor where the channel is made of nanocrystal sensitized graphene

Scientific approach: Our strategy is to use an intermediate band gap 2D material such as InSe. The value of the band gap is chosen so that the dark current will be reduced thanks to the moderate thermal activation of the carrier and at the same time the moderate band gap eases the charge transfer from the narrow band gap NC. Regarding the NC, we target to use HgSe NCs, which are self-doped, and present intraband absorption in the 3-5 μm range.

The second part of the project deals with the design of a light matter resonator. A first aspect of the design will be to enhance the light absorption in the spectral range where NCs absorb. A second constraint relates to the spatial localization of this absorption. We aim to localize the absorption near the 0D/2D interface. Such electromagnetic mode can be obtained from planar Fabry-Pérot for example, see a preliminary map below in **Figure 2**.

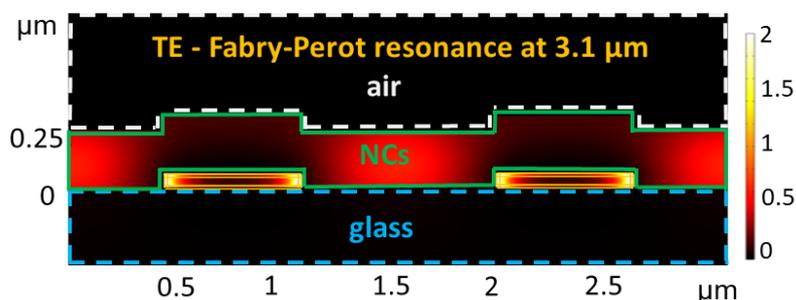


Figure 2 Absorption map for a NC film coupled to a metallic grating

Risks: A serious risk is to design a device with a slow time response. Gain that tends to enhance the photo-response is also responsible for slowing the response. The slow time response also strongly relates to carrier recombination within the NC. If ever it was the case a gate will be added and be used as an erase button. A second risk is that transport in the NC competes with the one in the 2D material due to the narrow band gap nature of the NC which makes the film quite conductive. To avoid that an additional fabrication step will be added to coat the contact with a dielectric to prevent conduction in the NC array.

- **Skills and coherence of team**

J. Biscaras is working on the transport properties of 2D materials^{iv}. At IMPMC, he developed the space charge doping technique (along with A. Shukla) and applied it to study phase transitions in 2D MoS₂, graphene, and high temperature superconductor 2D materials^{v vi}. The team has developed strong skills regarding the exfoliation of such 2D materials and their characterization for light sensors^{vii}.

E. Lhuillier is working on narrow band gap nanocrystals^{viii} and their device integration. His teams handle the growth of the nanomaterial as well as the infrared device characterization^{ix x}. More recently, the team has developed some expertise about the design and fabrication of sensors with controlled light matter coupling^{xi}

The two teams have complementary skills regarding the material and also regarding the experimental facilities (Raman and fundamental transport at IMPMC, fabrication and characterization at INSP).

2. Research plan with provisional calendar

The two laboratories are located close to each other, this will enable interaction on a daily basis, driven by project needs

First year: The first year will be focused on the training of the student to fabrication processes in INSP's cleanroom / exfoliation of the 2D material and on the structural (Raman and microscopy) and transport measurements. **Main expected result:** validation of the fabrication process and promoting student to independence.

Second year: the second year will be focused on the design of the light resonator. This period will include the electromagnetic design of the structure and its fabrication. μm -sized patterns are anticipated and thus e-beam lithography will be used. The device will be characterized in terms of responsivity, spectral response, time response, signal to noise ratio. This work will be systematically conducted as a function of temperature. **Main expected result:** demonstration of the light enhancement strategy

Third year: This period will be first focused on the characterization of the electronic structure of the hybrid structure and also on publication preparation. The last 6 months will be focused on thesis writing and defense preparation. **Main expected result:** revealing static and dynamics band electronic structure of the built device.

References

Ref from IMPMC - Ref from INSP - Other ref

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- ⁱ *Hybrid graphene–quantum dot phototransistors with ultrahigh gain*, G. Konstantatos et al, *Nature nanotechnology* 7, 363 (2012)
- ⁱⁱ *Hybrid 2D–0D MoS₂–PbS Quantum Dot Photodetectors*, D. Kufer et al, *Advanced Materials* 27, 176 (2015)
- ⁱⁱⁱ *Colloidal HgTe quantum dot/graphene phototransistor with a spectral sensitivity beyond 3 μm* , M.J. Grotevent et al, *Advanced Science* 8 (6), 2003360 (2021)
- ^{iv} *Multiple quantum criticality in a two-dimensional superconductor*, J. Biscaras et al, *Nature Materials* 12, 542 (2013)
- ^v *Superconductor-insulator transition in space charge doped one unit cell Bi_{2.1}Sr_{1.9}CaCu₂O_{8+x}*, F. Wang et al, *Nature Comm.* 12, 2926 (2021)
- ^{vi} *Onset of two-dimensional superconductivity in space charge doped few-layer molybdenum disulfide*, J. Biscaras et al, *Nature Comm.* 6, 8826 (2015)
- ^{vii} *A high performance graphene/few-layer InSe photo-detector*. Z. Chen et al, *Nanoscale*, 75981 (2015)
- ^{viii} *Mercury Chalcogenide Quantum Dots: Material Perspective for Device Integration*, C. Gréboval et al, *Chemical Reviews* 121, 3627 (2021)
- ^{ix} *Terahertz HgTe nanocrystals: beyond confinement*, N. Goubet et al, *Journal of the American Chemical Society* 140, 5033 (2018)
- ^x *Electroluminescence from Nanocrystals above 2 μm* , J. Qu et al, *Nature Photonics* 16, 38 (2022)
- ^{xi} *Ferroelectric Gating of Narrow Band-Gap Nanocrystal Arrays with Enhanced Light–Matter Coupling*, C. Gréboval et al, *ACS photonics* 8, 259 (2021)
- ^{xii} *Pushing absorption of perovskite nanocrystals into the infrared*, P. Rastogi et al, *Nano Letters* 20, 3999 (2020)
- ^{xiii} *Near Unity Absorption in Nanocrystal Based Short Wave Infrared Photodetectors using Guided Mode Resonators*; A. Chu et al, *ACS Photonics* 6, 2553 (2019)