

1.3.16 THz nanophotonic devices with 2D materials and 1D nanostructures

Bi-dimensional nano-materials and related heterostructures are establishing themselves as new photonic and electronic materials for the development of innovative devices, with huge potential in a variety of applications, ranging from saturable absorbers to optical modulators, from optical communication modules to spintronics, from near-field components to high-resolution sensing and fast tomography. Their peculiar band-structure and electron transport characteristics, which can be easily manipulated via layer-thickness control, suggest they could also form the basis for a new generation of high-performance devices operating in the Terahertz frequency range (1-10 THz) of the electromagnetic spectrum. We here review our latest achievements in THz nano-photonics and nano-electronics based on 2D nano-materials and combined heterostructures

The ability to convert light into an electrical signal with high efficiencies and controllable dynamics is a major need in photonics and optoelectronics. In the Terahertz (THz) frequency range, with its exceptional application possibilities in high-data-rate wireless communications, security, night-vision, biomedical or video-imaging and gas sensing, detection technologies providing efficiency and sensitivity performances that can be “engineered” from scratch, remain elusive. These key priorities prompted, in the last decade, a major surge of interdisciplinary research, encompassing the investigation of different technologies in-between optics and microwave electronics, different physical mechanisms and a large variety of material systems offering ad-hoc properties to target the expected performance and functionalities.

By exploiting the inherent electrical and thermal in-plane anisotropy of a flexible thin flake of black-phosphorus (BP), we recently devised plasma-wave, thermoelectric and bolometric nano-detectors with a selective, switchable and controllable operating mechanism. All devices operate at room-temperature at 0.3 THz and are integrated on-chip with planar nano-antennas, which provide remarkable efficiencies through light-harvesting in the strongly sub-wavelength device channel. The achieved selective detection (5-8 V/W responsivity) and sensitivity performances (signal-to-noise ratio of 500), are exploited to demonstrate the first concrete application of a phosphorus-based active THz device, for pharmaceutical and quality control imaging of macroscopic samples, in real-time and in a realistic setting.

Artificial semiconductor heterostructures played a pivotal role in modern electronic and photonic technologies, providing a highly effective means for the manipulation and control of carriers, from the visible to THz frequency range. Despite the exceptional versatility, they commonly require challenging epitaxial growth procedures, due to the need of clean and abrupt interfaces, which proved to be a major obstacle for the realization of room temperature (RT), high efficiency photonic devices, like sources, detectors or modulators, especially in the far-infrared. Inspired by the artificial semiconductor heterostructure architecture and the fascinating capabilities of van der Waals (vdW) heterostructures, we embedded a BP flake within a natural semiconductor heterostructure formed by multilayered hexagonal boron nitride (hBN) crystals to devise hBN/BP/hBN heterostructured THz photodetectors having high optical response, and an extremely good time-dependent electrical stability. The achieved selective detection (40 V/W responsivity) and sensitivity performances (signal-to-noise ratio of 10000), has been exploited for real-time THz imaging of biological samples in a realistic setting and for application of process and quality control.

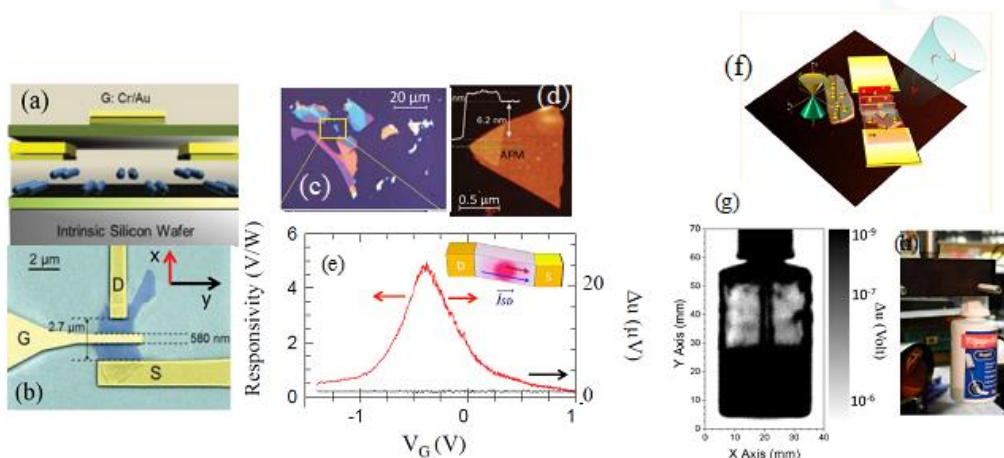


Figure 1. (a) Sketch of a black phosphorus (BP) photodetector device structure (vertical section). (b) False colors SEM image of the BP-based FET. The channel length (L_c) is $2.7 \mu\text{m}$, the gate length (L_g) is 530 nm . (c) Optical image of exfoliated flakes of BP. (d) Atomic force microscopy topographic image of an individual BP flake with thickness 6.2 nm . A topographic line profile, acquired along the dashed green line is shown. (e) Gate bias dependence of the experimental room temperature responsivity. The red line was measured by impinging the THz beam on the detector surface; the black line was measured while blanking the beam with an absorber (considering unaltered the incident power). Inset: schematics of the over-damped plasma-wave dynamics. (f) Schematic sketch of a THz detection process mediated by topological insulator surface states. (g) Room temperature, large area THz imaging obtained while impinging the 332.6 GHz radiation a topological insulator THz detector, mounted on a XY stage, with an acquisition time of 20 ms/pixel . For visible light illumination the contents cannot be seen, either by naked eye or by the CCD camera used to take the picture. The detection of THz transmitted radiation gives information about the jar content. (h) Photograph of the glue jar.

As a very intriguing alternative, we explored topological insulators (TIs), which represent a novel quantum state of matter, characterized by edge or surface-states, showing up on the topological character of the bulk wave-functions. Allowing electrons to move along their surface, but not through their inside, they emerged as an intriguing material platform for the exploration of exotic physical phenomena, somehow resembling the graphene Dirac-cone physics, as well as for exciting applications in optoelectronics, spintronics, nanoscience, low-power electronics and quantum computing. Investigation of topological surface states (TSS) is conventionally hindered by the fact that, in most of experimental conditions, the TSS properties is mixed up with those of bulk-states. We devised a novel tool to unveil TSS and to probe related plasmonic effects. By engineering $\text{Bi}_2\text{Te}_{(3-x)}\text{Se}_x$ stoichiometry, and by gating the surface of nanoscale field-effect-transistors, exploiting thin flakes of $\text{Bi}_2\text{Te}_{2.2}\text{Se}_{0.8}$ or Bi_2Se_3 , we recently provided the first demonstration of room-temperature THz detection mediated by over-damped plasma-wave oscillations on the “activated” TSS of a $\text{Bi}_2\text{Te}_{2.2}\text{Se}_{0.8}$ flake.

Uncooled THz photodetectors (PDs) combining such a high sensitivity (noise equivalent power (NEP) $< \text{nW}/\text{Hz}^{1/2}$) with fast response times, over a broad (0.1 - 10 THz) frequency range are needed for applications in high-resolution spectroscopy (precisions of 10^{-11}), metrology, quantum information, security, imaging, optical communications. However, present THz receivers cannot provide

the required balance between sensitivity, speed, operation temperature and frequency range.

To address this goal, we developed THz-frequency detectors exploiting hBN/graphene/hBN heterostructures that exploit the photo-thermoelectric effect. The core structure relies on a novel architecture that employs a dual-gated, dipolar antenna with a gap of 100 nm. We demonstrate that this new detector has excellent sensitivity, with a noise-equivalent power of $80 \text{ pW/Hz}^{1/2}$ at room temperature, a response time below 30 ns (setup-limited), a high dynamic range (linear power dependence over more than 3 orders of magnitude) and broadband operation (measured range 1.8 - 4.2 THz, antenna-limited), which fulfills a combination that is currently missing in the state of the art (Figs 2a-b).

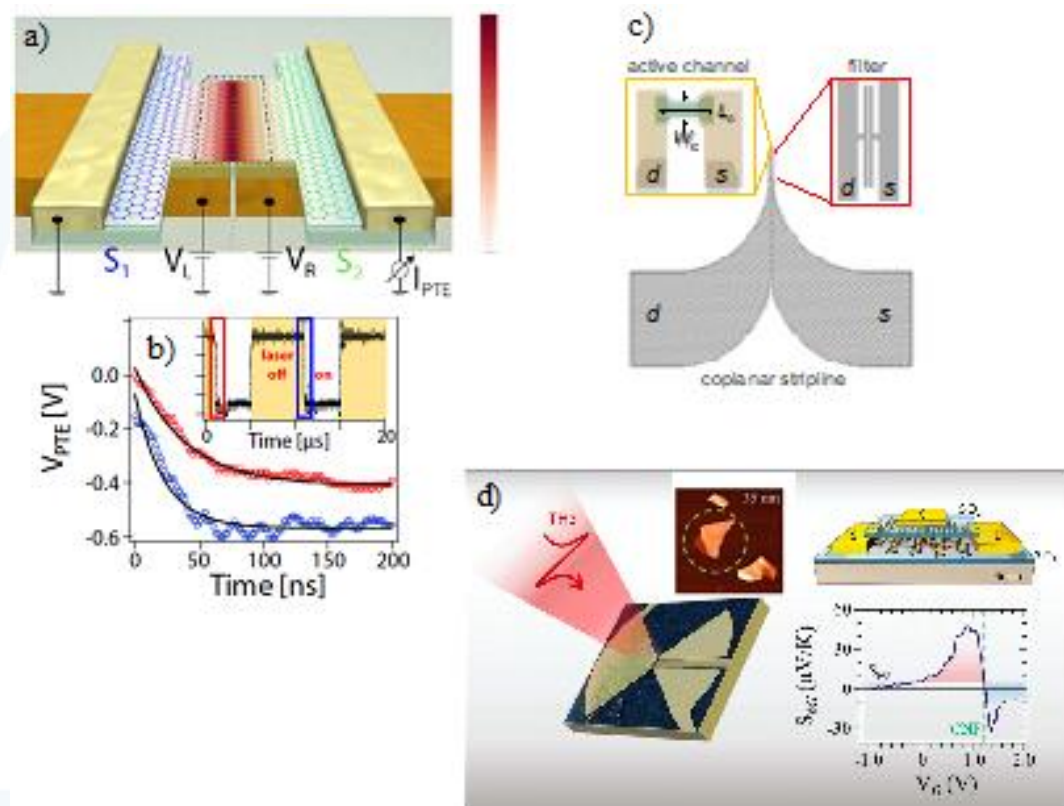


Figure 2. (a) Schematic representation (right; not to scale) of the antenna-integrated pn-junction graphene photodetector; (b) Results of the pulsed laser experiment, where the photocurrent was amplified by a fast current pre-amplifier (Femto) and the data were acquired with a fast oscilloscope. The inset shows how the photovoltage V_{PTE} follows the switching of the pulsed laser; (c) On-chip RF components. The S and D electrodes are shaped in a CPS geometry. Inset (left): the shape of the active LMH channel (green area) guarantees a lower contact resistance with respect to a rectangular geometry. The S and D contacts have a thickness of 45 nm in vicinity of the GFET channel (yellow areas) and a thickness of 140 nm far from the GFET channel. Inset (right): planar low-pass filter, with cut-off frequency 300 GHz; (d) Terahertz detection activated by photo-thermoelectric effect in selenium-doped thin (35 nm) black phosphorus flakes.

We also demonstrate that by integrating our RT THz nano-receivers with lithographically patterned high-bandwidth ($\sim 100 \text{ GHz}$) chips, we can further improve the detection speed to hundred ps response times, preserving the high sensitivity (Fig. 2c). Remarkably, this can be achieved with various antenna and

transistor architectures (single gate, dual gate) for any frequency in the 0.1-10 THz range, thus paving the way for the design of ultrafast graphene arrays in the far infrared, opening concrete perspective for targeting the aforementioned applications

If graphene can be an interesting material system for THz oriented applications due to its high mobility and gapless nature, the inherent in-plane anisotropy of BP, combined with the tunable bandgap, makes it an appealing and intriguing alternative for many applications in the far-infrared. We therefore exploited the intrinsic chemical stability of thin flakes of Se-doped BP, combined with the strong electrical and thermal anisotropy and the possibility to control, via its thickness, the energy gap to develop highly sensitive room-temperature photodetectors at high frequencies (3.4 THz) with state-of-the-art performances and different layer thicknesses (Fig.2d).

We also pioneered the use semiconductor nanowires (NW) as building block for implementing rectifying diodes or detectors that could be well operated into the THz, thanks to their typical achievable attofarad-order capacitance. We therefore devised 1D InAs or InAs/InSb NW-based field effect transistors (FETs) exploiting novel morphologies and/or material combinations effective for addressing the goal of a semiconductor plasma-wave THz detector array technology. We demonstrated room-temperature operation in the 0.3-3 THz range with over 100V/W responsivity and $< 1\text{ nW/Hz}^{1/2}$ noise equivalent powers.

In parallel, we also worked on the development of novel passive and active optical components. THz saturable absorbers (SA) can be extremely appealing in combination with QCLs to passively mode-lock these micro-sources. We developed flexible THz SAs by transfer coating and inkjet printing single and few-layer graphene films prepared by liquid phase exfoliation of graphite, achieving a record transparency modulation of 80% at 3 THz (Fig. 3), almost one order of magnitude larger than that reported to date at THz frequencies.

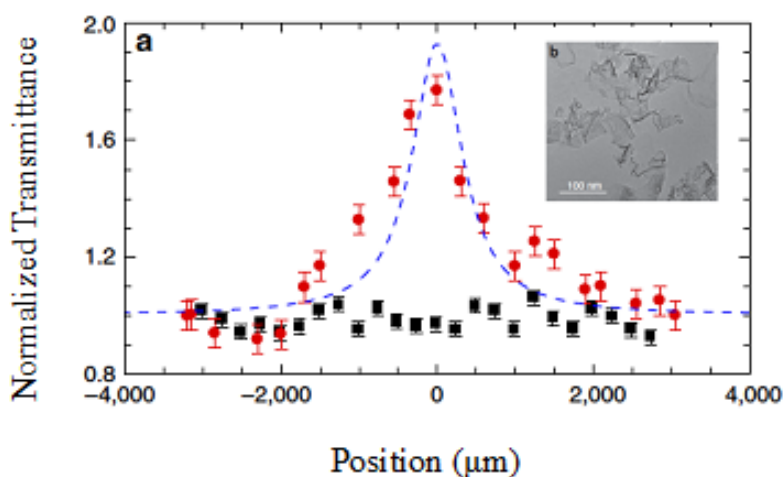


Figure 3. (a) z-scan normalized transmittance of a water-based graphene saturable absorber probed with a 3.4 TH QCL (b) Transmission electron microscopy images of few-layer graphene flakes from water based inks.

References

- [1] M.S. Vitiello, *Nanodevices at terahertz frequency based on 2D materials*, Journal of Physics: Materials **3** (1), 014008 (2019).
- [2] S. Castilla, B. Terrés, M. Autore, L. Viti, J. Li, A.Y. Nikitin, I. Vangelidis, K. Watanabe, T.

- Taniguchi, E. Lidorikis, M.S. Vitiello, R. Hillenbrand, K.-J. Tielrooij, F.H.L. Koppens, *Fast and sensitive terahertz detection using an antenna-integrated graphene pn junction*, *Nano Letters* **19**, 2765 (2019).
- [3] L. Viti, A. Politano, K. Zhang, M.S. Vitiello, *Thermoelectric terahertz photodetectors based on selenium-doped black phosphorus flakes*, *Nanoscale* **11**, 1995 (2019).
- [4] A. Agarwal, M.S. Vitiello, L. Viti, A. Cupolillo, A. Politano, *Plasmonics with two-dimensional semiconductors: from basic research to technological applications*, *Nanoscale* **10**, 8938 (2018).
- [5] C. Liu, L. Wang, X. Chen, A. Politano, D. Wei, G. Chen, W. Tang, W. Lu, A. Tredicucci, *Hot-Carrier-Driven Photodetection: Room-Temperature High-Gain Long-Wavelength Photodetector via Optical-Electrical Controlling of Hot Carriers in Graphene*, *Advanced Optical Materials* **6**, 1870093 (2018).
- [6] V. Bianchi, T. Carey, L. Viti, L. Li, E.H. Linfield, A.G. Davies, A. Tredicucci, D. Yoon, P.G. Karagiannidis, L. Lombardi, F. Tomarchio, A.C. Ferrari, F. Torrisi, M.S. Vitiello, *Terahertz saturable absorbers from liquid phase exfoliation of graphite*, *Nature Communications* **8**, 1 (2017).
- [7] L. Viti, A. Politano, M.S. Vitiello, *Black phosphorus nanodevices at terahertz frequencies: Photodetectors and future challenges*, *APL Materials* **5**, 035602 (2017).
- [8] A. Politano, L. Viti, M.S. Vitiello, *Optoelectronic devices, plasmonics, and photonics with topological insulators*, *APL Materials* **5** (3), 035504 (2017).
- [9] A. Politano, M.S. Vitiello, L. Viti, D.W. Boukhvalov, G. Chiarello, *The role of surface chemical reactivity in the stability of electronic nanodevices based on two-dimensional materials "beyond graphene" and topological insulators*, *Flat Chem* **1**, 60 (2017).
- [10] A. Politano, M.S. Vitiello, L. Viti, J. Hu, Z. Mao, J. Wei, G. Chiarello, D.W. Boukhvalov, *Unusually strong lateral interaction in the CO overlayer in phosphorene-based systems*, *Nano Research* **9**, 2598 (2017).
- [11] L. Viti, J. Hu, D. Coquillat, A. Politano, C. Consejo, W. Knap, M.S. Vitiello, *Heterostructured hBN-BP-hBN Nanodetectors at Terahertz Frequencies*, *Advanced Materials* **28**, 7390 (2016).
- [12] A.M. Kadykov, C. Consejo, M. Marcinkiewicz, L. Viti, M.S. Vitiello, S.S. Krishtopenko, S. Ruffenach, S.V. Morozov, W. Desrat, N. Dyakonova, W. Knap, V.I. Gavrilenko, N.N. Mikhailov, S.A. Dvoretzky, F. Teppe, *Observation of topological phase transition by terahertz photoconductivity in HgTe based transistors*, *Physica Status Solidi C* **13** (7-9), 534 (2016).
- [13] L. Viti, J. Hu, D. Coquillat, A. Politano, W. Knap, M.S. Vitiello, *Efficient Terahertz detection in black-phosphorus nano-transistors with selective and controllable plasma-wave, bolometric and thermoelectric response*, *Nature Scientific Reports* **6**, 20474 (2016).
- [14] L. Viti, D. Coquillat, A. Politano, K.A. Kokh, Z.S. Aliev, M.B. Babanly, O.E. Tereshchenko, W. Knap, E.V. Chulkov, M.S. Vitiello, *Plasma-wave terahertz detection mediated by topological insulators surface states*, *Nano Letters* **16** (1), 80 (2016).
- [15] A. Politano, V.M. Silkin, I.A. Nechaev, M.S. Vitiello, L. Viti, Z.S. Aliev, M.B. Babanly, G. Chiarello, P.M. Echenique, E.V. Chulkov, *Interplay of Surface and Dirac Plasmons in Topological Insulators: The Case of Bi₂Se₃*, *Physical Review Letters* **115**, 216802 (2015).
- [16] A.M. Kadykov, F. Teppe, C. Consejo, L. Viti, M.S. Vitiello, S.S. Krishtopenko, S. Ruffenach, S.V. Morozov, M. Marcinkiewicz, W. Desrat, N. Dyakonova, W. Knap, V.I. Gavrilenko, N.N. Mikhailov, S.A. Dvoretzky, *Terahertz detection of magnetic field-driven topological phase transition in HgTe-based transistors*, *Applied Physics Letters* **107**, 152101 (2015).
- [17] L. Viti, J. Hu, D. Coquillat, W. Knap, A. Tredicucci, A. Politano, M.S. Vitiello, *Black phosphorus terahertz photodetectors*, *Advanced Materials* **27**, 5567 (2015).
- [18] F. Bianco, D. Perenzoni, D. Convertino, S.L. De Bonis, D. Spirito, M. Perenzoni, C. Coletti, M.S. Vitiello, A. Tredicucci, *Terahertz detection by epitaxial-graphene field-effect-transistors on silicon carbide*, *Applied Physics Letters* **107**, 131104 (2015).
- [19] F. Bianco, V. Miseikis, D. Convertino, J.-H. Xu, F. Castellano, H.E. Beere, D.A. Ritchie, M.S. Vitiello, A. Tredicucci, C. Coletti, *THz saturable absorption in turbostratic multilayer graphene on silicon carbide*, *Optics Express* **23**, 11632 (2015).
- [20] M.S. Vitiello, L. Viti, D. Coquillat, W. Knap, D. Ercolani, L. Sorba, *One dimensional semiconductor nanostructures: An effective active-material for terahertz detection*, *APL Materials* **3** (2), 026104 (2015).
- [21] L. Romeo, D. Coquillat, E. Husanu, D. Ercolani, A. Tredicucci, F. Beltram, L. Sorba, W. Knap, M.S. Vitiello, *Terahertz photodetectors based on tapered semiconductor nanowires*, *Applied Physics Letters* **105** (23), 231112 (2014).

- [22] F.H.L. Koppens, T. Mueller, P. Avouris, A.C. Ferrari, M.S. Vitiello, M. Polini, *Photodetectors based on graphene, other two-dimensional materials and hybrid systems*, Nature Nanotechnology **9**, 780 (2014).
- [23] L. Viti, D. Coquillat, D. Ercolani, L. Sorba, W. Knap, M.S. Vitiello, *Nanowire Terahertz detectors with a resonant four-leaf-clover-shaped antenna*, Optics Express **22**, 8996 (2014).
- [24] M. Ravaro, M. Locatelli, L. Viti, D. Ercolani, L. Consolino, S. Bartalini, L. Sorba, M.S. Vitiello, P. De Natale, *Detection of a 2.8 THz quantum cascade laser with a semiconductor nanowire field-effect transistor coupled to a bow-tie antenna*, Applied Physics Letters **104** (8), 083116 (2014).
- [25] D. Spirito, D. Coquillat, S.L. De Bonis, A. Lombardo, M. Bruna, A.C. Ferrari, V. Pellegrini, A. Tredicucci, W. Knap, M.S. Vitiello, *High performance bilayer-graphene terahertz detectors*, Applied Physics Letters **104**, 061111 (2014).