BI-DIMENSIONAL MATERIALS FOR THz FREQUENCY NANODEVICES

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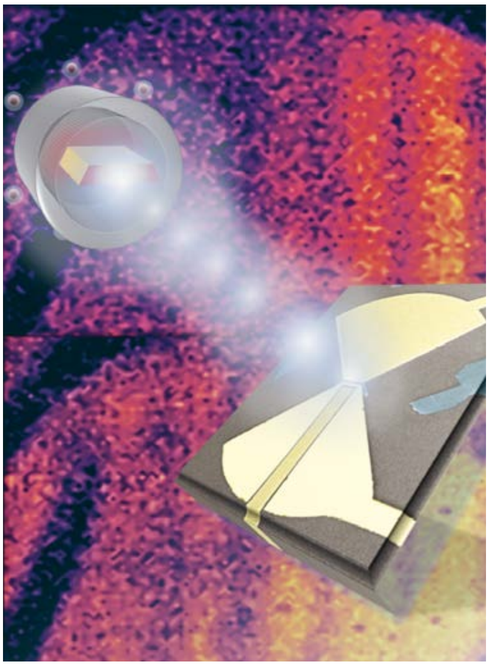

Although artificial semiconductor heterostructures have long been the core material system for the generation, detection and manipulation of carriers, at TeraHertz (THz) frequencies, the discovery of graphene and the related intriguing abilities have triggered an unprecedented interest in inorganic two-dimensional (2D) materials, as black phosphorus and boron nitride, amongst many others. They offer a unique platform for developing efficient devices, without the need of lattice matching, and with a variety of physical properties, that can be engineered from scratch, exploiting the material structures, the layer thickness or their inherent anisotropy.

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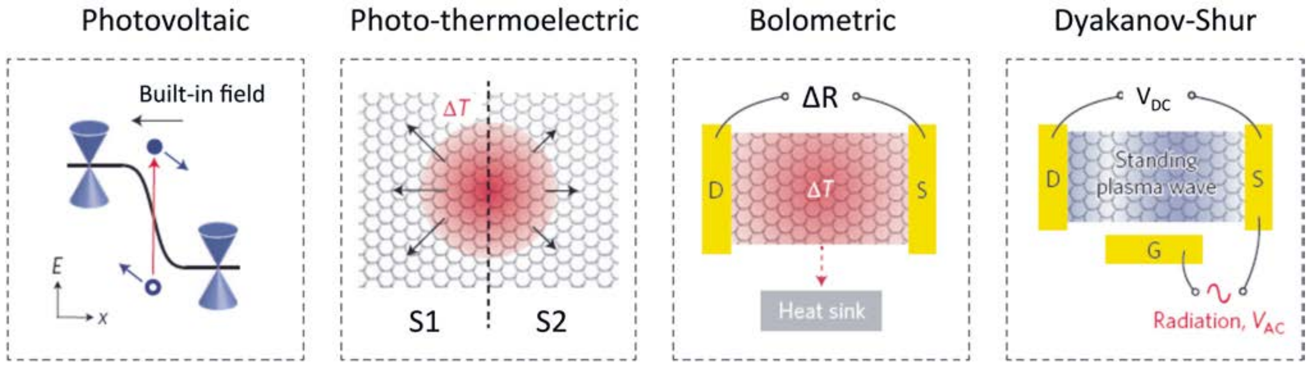
The past decade has witnessed the rise of graphene — a single atomic sheet of graphite — as one of the most studied condensed matter systems. The success of graphene has been followed by a wealth of recent research efforts to study other forms of atomically thin two-dimensional (2D) materials, such as hexagonal boron nitride (hBN) and black phosphorus (BP). This surge of interest in 2D materials is attributed to their exceptional electronic, optical, and magnetic properties and also to their unique amenability to layer-by-layer assembly. That is, multiple different 2D materials can be combined layer by layer into various van der Waals (vdW) heterostructures.

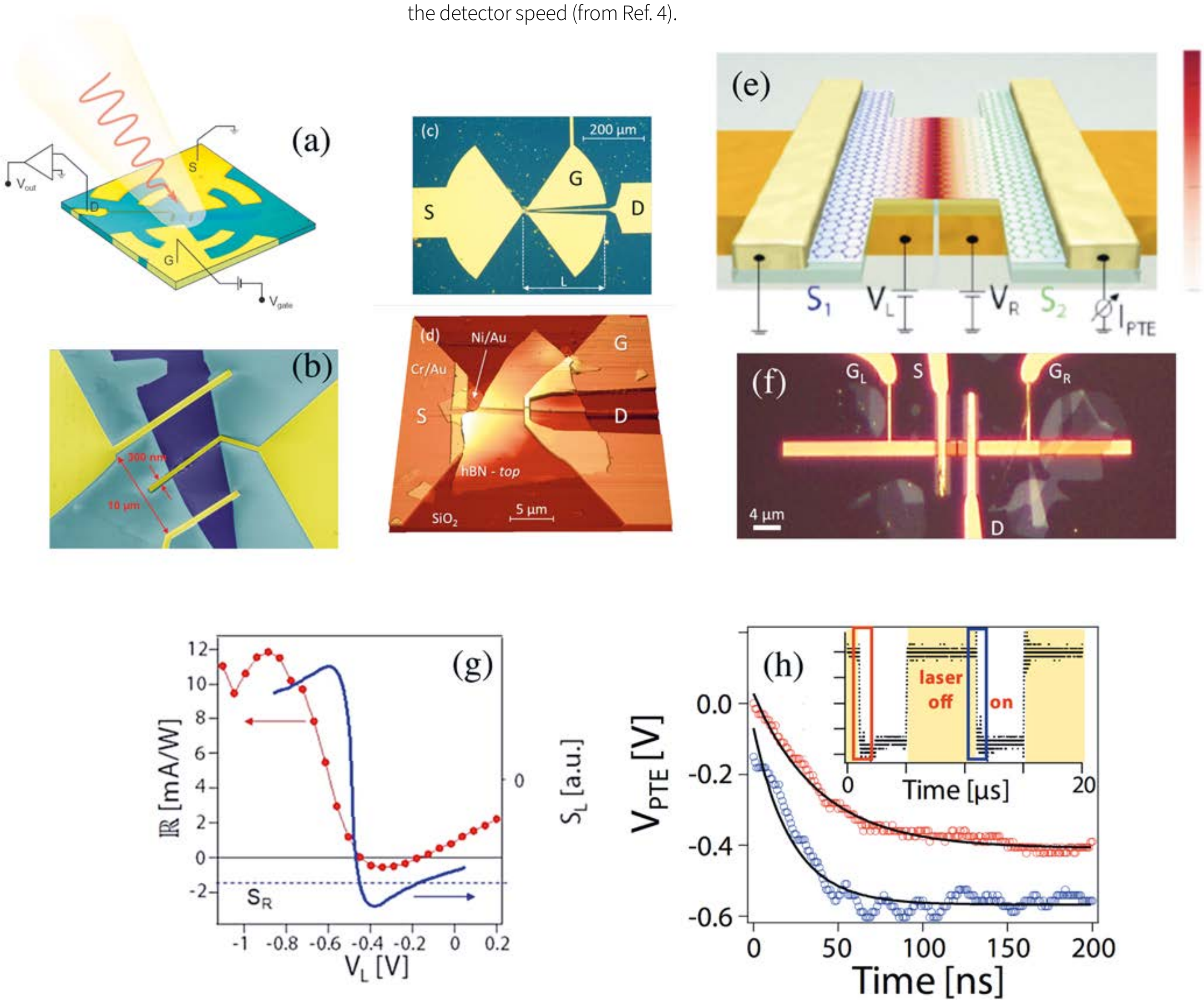
These engineered quantum structures are of both basic and applied interest. They indeed display an extraordinary technological potential for engineering nano-electronic and nano-photonic devices and components; they also provide an intriguing platform for fundamental investigations at the nanoscale, through the exploitation of their confined electronic systems. Intriguingly, the possibility of hybridizing collective electronic motion with light in so-called surface polaritons has also made these materials a versatile platform for extreme light confinement and tailored nanophotonics. Finally, being fully compatible with a wide range of substrates including flexible and transparent ones, if placed on chip with flat integrated optical circuits, they can maximize interaction with light, therefore optimally utilizing their versatile properties for a wealth of applications in transformational optics and high-resolution tomography.

The family of 2D materials is now gaining a renewed interest in more unexplored frequency domains, like the THz) frequency range (0.1-10 THz, 3000–30 µm). • • •



△ Figure 1.
Schematic representation of the 4 photocurrent generation mechanisms discussed in the main text (from Ref. 1).

▽ Figure 2.
(a) Schematic representation of the graphene FET overdamped plasma wave detector layout with a patterned log-periodic nano-antenna and (b) false-color SEM micrograph of the graphene flake embedded in the FET channel (from Ref. 2). (c) Optical microscopy image of an hBN/BP/hBN FET with a split bow-tie antenna and d) AFM tomographic image of the top-gate field effect transistor (from Ref. 3). (e) Schematic representation (right; not to scale) of the antenna-integrated pn-junction device and (f) related optical microscope image of the fabricated device; (g) Photoresponse as a function of voltages applied to the two antenna branches/gates at 2.52 THz; The blue line represents the calculated Seebeck coefficient; (h) evolution of the thermoelectric voltage as a function of the time from which we extracted the detector speed (from Ref. 4).



Dynamical phenomena (scattering, recombination, and tunneling) in 2D materials indeed typically occur on a time scale of picosecond, *i.e.* at THz frequencies. Graphene and related materials can therefore offer an intriguing perspective for engineering novel THz electronic or photonic devices. Here, I will review some examples of 2D material based THz frequency nano-devices.

A) GRAPHENE AND BLACK-PHOSPHORUS PHOTODETECTORS

Photodetection of light relies on the conversion of photons into a stable electrical signal. In a nanomaterial, and at THz frequencies, such a process can be accomplished by several different physical mechanisms like photo-thermoelectric, photovoltaic, galvanic, bolometric, plasma-wave rectification or can occur through a combination of different effects [1].

In the last few years, 2D materials demonstrated to be an ideal building block for devising THz photo-detectors. As a prototypical example, graphene exhibits ultrafast carrier dynamics, wavelength-independent absorption, tunable optical properties *via* electrostatic doping, and high-mobility, which enables ultrafast conversion of photons or plasmons to electrical currents or voltages. As a major distinctive characteristic, graphene is gapless, allowing charge carrier generation by light absorption over a very wide energy spectrum, while always conducting a significant amount of electricity, meaning that its inherent "leakage" can partially affect the device efficiency.

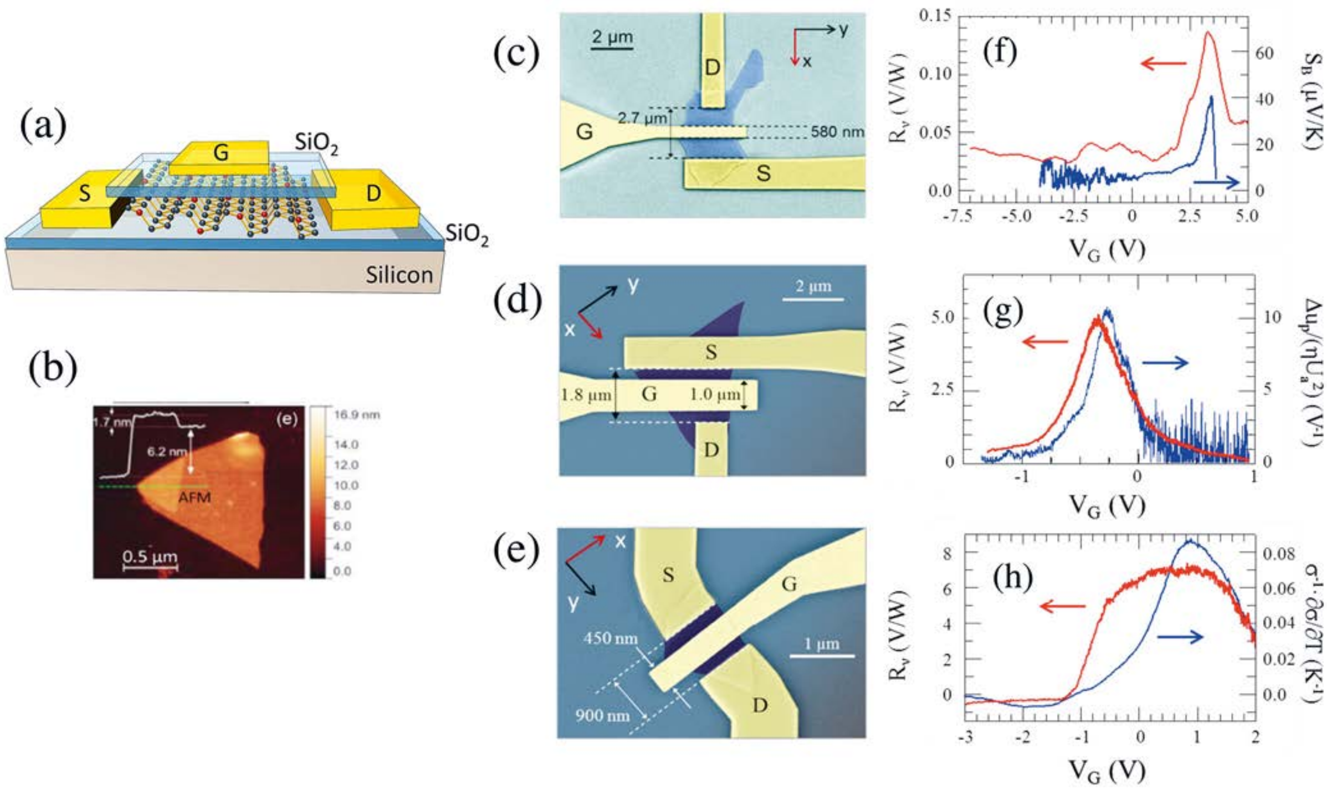
Conversely, BP behaves like a semiconductor, meaning that it only conducts electricity whenever the electrons absorb enough energy through heat, light, and other means. Depending on its specific layer thickness and related specific band structure, and due to its inherently high anisotropy, it can allow engineering the detection dynamics from scratch, allowing highly efficient light detection.

Usually, field effect transistors (FETs) are the most commonly employed architecture for devising THz frequency photodetectors. FETs indeed provide some clear advantages at those frequencies, namely the inherent scalability and the combination of a fast response and high frequency operation (up to 22 THz), very differently from Schottky diodes, whose performances are strongly affected by parasitic capacitances and usually show a dramatic cutoff above 1 THz.

The rich physics involved in 2D materials can be exploited in a FET to engineer the detection dynamics from scratch, playing with the geometrical symmetry. A bolometer can be engineered by exploiting the variation of the channel conductance induced by the homogeneous heating of the channel, either by applying a source-to-drain bias, or, by symmetrically feeding the THz radiation *via* an on-chip integrated symmetric resonant antenna. Conversely, photo-thermoelectric and plasma-wave rectification both require a certain degree of asymmetry in the detector structure. In the first case, this is achieved by inducing a temperature gradient along the FET channel, for example by creating a pn-junction along the FET channel; in the second case, plasma-waves can be rectified inside the transistor when the THz field is coupled asymmetrically, for example between source and gate electrodes.

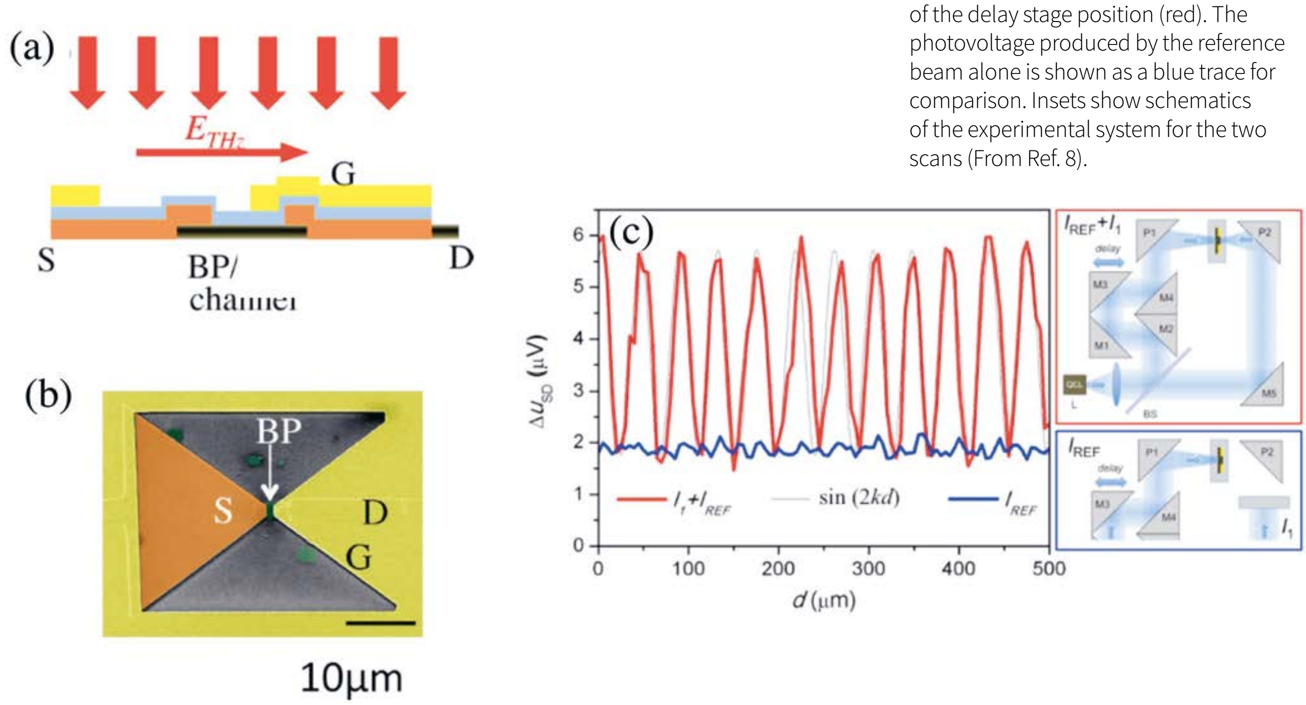
Following the first demonstrations of THz graphene photodetectors operating in the overdamped plasma waves resistive self-mixing regime [2], several architectures have been proposed and implemented [1], including graphene bolometers, ballistic rectifiers [3], and fast photo-thermoelectric sensors, involving a combination of high-mobility hBN-encapsulated graphene and small-area graphene pn-junctions [4].

State of the art performances include room-temperature (RT) operation over the 1.8–4 THz range • • •



△ **Figure 3.** (a) Schematic representation of the black phosphorus FET detector; (b) AFM image of the intra-channel thin flake of BP (From Ref. 5) (c-e) False-color SEM micrographs of a set of BP FETs with the BP flake oriented along the armchair direction (c), the direction at 45° in between (d), or along the zig-zag direction (e); (f) gate voltage dependence of the responsivity (left vertical axis) of the thermoelectric detector of panel (c) and gate voltage dependence of the extrapolated Seeback coefficient (right vertical axis) (From Ref. 6); (g) gate voltage dependence of the responsivity (left vertical axis) of the over-damped plasma wave detector of panel (d) and gate voltage dependence of the predicted theoretical photovoltage (right vertical axis) (From Ref. 6); (f) gate voltage dependence of the responsivity (left vertical axis) of the bolometer of panel (e) and gate voltage dependence of the extrapolated bolometric coefficient (right vertical axis) (From Ref. 6).

▽ **Figure 4.** THz near-field probe with an embedded nano-detector. (a) Electric field E (red arrow) of the THz wave induces oscillating field between the source (S) and gate (G) electrodes (From Ref. 8). (b) Scanning electron microscopy (SEM) image of a near-field probe with a 20 µm aperture and an embedded thin (14 µm) flake of black phosphorus (BP) (From Ref. 8). (c) Coherent detection using a near-field probe with an embedded NW detector; photovoltage plot as a function of the delay stage position (red). The photovoltage produced by the reference beam alone is shown as a blue trace for comparison. Insets show schematics of the experimental system for the two scans (From Ref. 8).



> Dynamical phenomena (scattering, recombination, and tunneling) in 2D materials indeed typically occur on a time scale of picosecond, *i.e.* at THz frequencies.

with NEPs = 80 pW/Hz$^{1/2}$, a dynamic range extending over four decades [4] and 890 ps response time, significantly above any other RT receiver commercially available and demonstrated so far.

As an alternative option, thin flakes of exfoliated BP can be employed in an antenna-coupled THz nanodetector to selectively activate a specific detection process [2,5,6]. The inherent electrical and thermal in-plane anisotropy of BP can be indeed exploited to selectively control the detection dynamics in the BP channel, with state-of-the-art performances. RT operation with ~ 20000 signal-to-noise ratio [2], NEPs ≤10^{-9} W/Hz$^{1/2}$), responsivities > 10 V/W [6], and response times of a few µs [7].

B) NEAR FIELD OPTICAL PROBES

Near-field imaging with sub-10-nm resolution is usually achieved by scattering near field optical spectroscopy (*s-SNOM*), exploiting an atomic force microscope (AFM) tip which converts the incident light into strongly concentrated fields at the tip apex (nanofocus) to locally excite molecular vibrations, plasmons or phonons in the sample. The spatial resolution is thus determined by the tip apex size, but limited by the weak scattering efficiency of the tip.

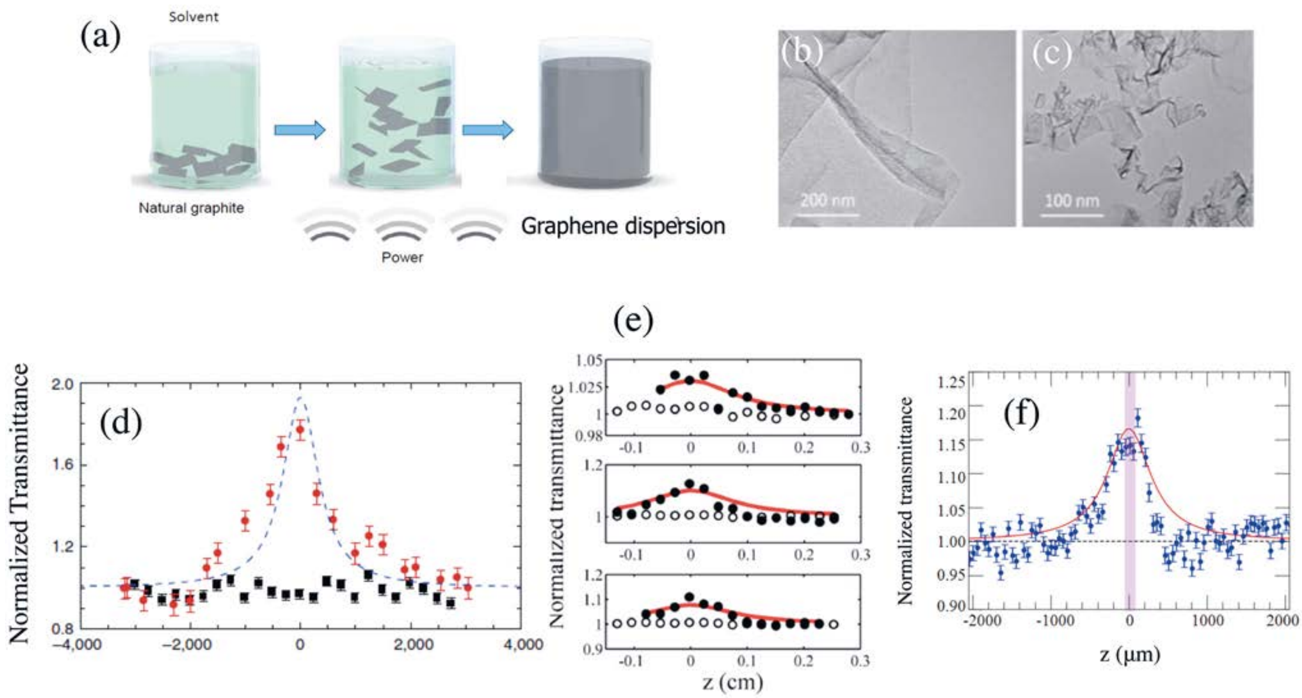
In the THz range, the scattering efficiency of AFM tips is prohibitively low, demanding the use of powerful gas lasers combined with cooled bolometers, or THz time-domain spectroscopy (THz-TDS) systems, which can detect very weak scattered fields, but with limited spectral resolution and slow image acquisition.

At the other end of the spectrum of length scales targeted by THz microscopy, are applications requiring mapping THz absorption properties and THz field distributions in large area samples. The s-SNOM approach is not appropriate for these applications, due to AFM instabilities arising during scans of rough or soft surfaces at the scan-speeds required for large-area imaging. As such, THz microscopy approaches that can address these applications are based on near-field probes with integrated sub-wavelength size THz detectors (a-SNOM). In this latter case, the resolution is limited by the strong reduction of light transmission (*T*) through an aperture (dimension a), according to the Bethe and Bouwkamp law, that follow the power law: $T \sim a^6$. Usually, although the field conveyed at the aperture includes both the transmitted and the reflected beam components, only transmitted signals are collected, meaning that the evanescent fields remain practically undetected.

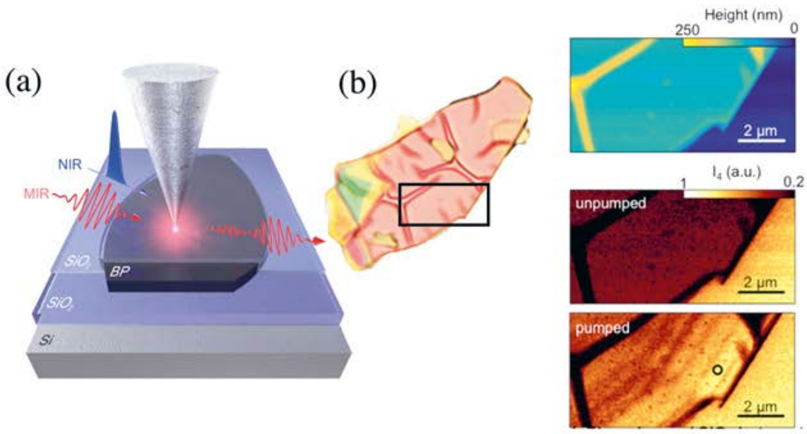
Recently, a novel near-field THz probe concept, in which the evanescent THz field is converted into a detectable electrical signal at the nanoscale has been proposed [8]. To do that, thin crystalline flakes of BP have been integrated into the evanescent field region of a trapezoidal shaped sub-wavelength aperture, simultaneously acting as a probe and as an evanescent field -sensitive photodetector [8]. This allowed realizing, for the first time, coherent THz detection (amplitude and phase) within the a-SNOM probe [8], eliminating the need for a THz TDS system or an external FTIR spectrometer.

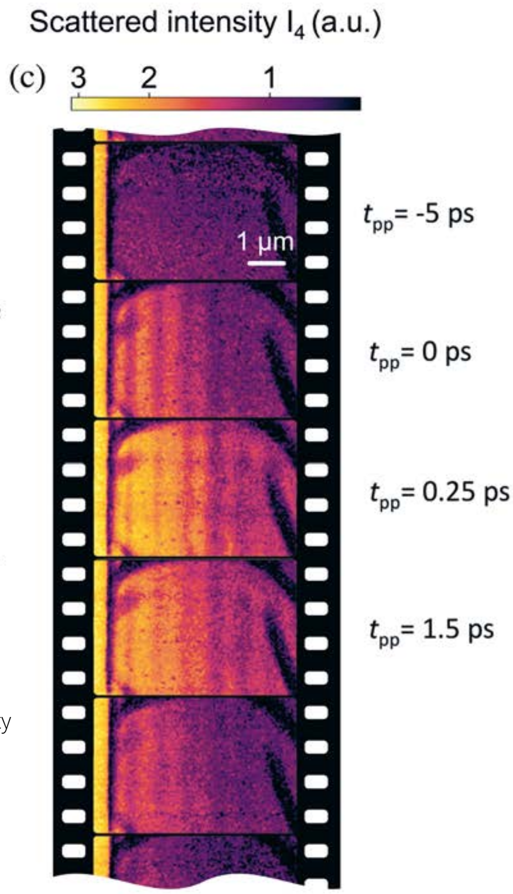
C) SATURABLE ABSORBERS

Semiconductor saturable-absorbers are poorly suitable for applications at THz frequencies since the photon energy is smaller than the • • •



△ Figure 5.
(a) Schematics of liquid phase graphene exfoliation; (b-c) Transmission electron microscopy images of (b) single-layer graphene flakes from ethanol based and (c) few-layer graphene flakes from water based inks (From Ref. 9). (d-f) z-scan normalized transmittance traces of (d) the water-based graphene saturable absorber (From Ref. 9), of (e) multi-layer graphene grown on the carbon-face of silicon carbide having different layer numbers (N=20, N = 80, N=90, from top to bottom) (From Ref. 10, © OSA), and (f) 50 layer graphene films, grown via CVD on Nickel (From Ref. 7),. The red lines in panel (e-f) and the dashed blue line in panel (d) are the fit curves assuming the simple two-level saturable absorber model.





△ Figure 6.
(a) Schematic of the set-up. An ultrafast near-infrared pump pulse is focused onto a SiO_2/BP/SiO_2 heterostructure. The sample is probed in the near field with a mid-infrared pulse coupled to a sharp metallic tip. Right: optical image of a single BP flake capped with SiO_2. (b) Spatial dependence of the scattered near-field intensity with and without pump excitation. Top: unpumped sample. Bottom: sample at a delay time between the pump and the probe tpp= 250 fs, after excitation by a near-infrared pulse with a fluence of 1 mJ cm^{-2}. The black circle indicates the position where the data shown in Fig. 2c were taken. All scale bars, 2 µm. (c) Scattered near-field intensity images of the heterostructure plotted for eight different delay times between the pump and probe pulses (From Ref. 11).

ADVERTORIAL

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GLOphotonics (or GLO for short) is a French start-up based in Limoges. It was founded by Fetah Benabid in Bath, UK, in 2008, then transferred to, and re-incubated in Limoges, France, in 2011. GLO is a pioneering market-player in the fabrication and commercialisation of its proprietary hollow-core photonic-crystal-fiber (HCPCF) and photonic microcell™ (PMC).

HCPCF is an optical fiber that stands out from conventional optical fibers by its light guidance, which allows guiding light in a hollow channel. We distinguish two types of guidance mechanisms in HCPCF. The first is called "Photonic Band Gap" and the second is called "Inhibited Coupling". A PMC is a photonic device, which consists of a length of HCPCF filled with a given gas and sealed in a hermetic and controllable fashion. HCPCF and PMC have outstanding key enabling features for a large number of applications, such as optical frequency conversion, ultra-high power pulsed laser beam delivery, laser pulse compression, sensing and precision-timing.

Based on HCPCF and PMC technology, GLO provides the market with a diverse portfolio of components, sub-systems, and technology solutions. In laser beam delivery, GLO offers a large range of products spanning from a tailored HCPCF to highly sophisticated ultra-short laser beam delivery products that include gas handling, thermal management, laser beam pointing stabilization and integration into systems.

FastLas is a unique product family to compress sub-picosecond laser pulses by a factor as high as 30, and achieves pulses as short as ~10 fs. Fastlas technology stands out with its large dynamic in laser pulse energy and wavelength.

Comblas is a light source product family that comprises sources emitting broadband combs spanning from UV (240 nm) to IR. Comblas is an ideal alternative to supercontinuum sources when high spectral power density is required, or to develop light source at exotic wavelength ranges.

In sensing and precision-timing, GLO develops specific PMC as highly efficient and compact spectroscopy cells.

GLO takes pride in counting a large and diverse customers base, from individual scientists to tier 1 industrial companies, and in addressing their requirements with the same level of commitment, and in providing them with unique and high added-value solutions.

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semiconductor band gap and the free carrier absorption losses very high.

Graphene is a potential candidate for crafting saturable absorbers at THz frequencies, thanks to its fast carrier dynamics (< 100 fs), large absorption of incident light (2.3% per layer), the possibility to saturate this absorption in a broad spectral range with relatively low incident power, and its tunable modulation depth. Its optical conductivity being mainly determined by intraband transitions (with a further interband relaxation dynamic, due to hot phonons cooling), graphene optical absorption can be easily modulated by electrical/optical control of its Fermi level. Furthermore, large-area, low-cost, single or multi-layer graphene can be easily grown and integrated in THz laser systems.

THz saturable absorption has been recently demonstrated in graphene [9,7]. By transfer coating and ink-jet printing 50 layers (randomly distributed) of graphene films prepared by liquid phase exfoliation of graphite and through a combination of open-aperture z-scan experiments and Fourier transform infrared (FTIR) spectroscopy, 80% transparency modulation at 3.4 THz has been reported [9]. Despite this technology is very appealing for intracavity embedding, since the graphene is printable and flexible, the thickness uniformity along a large surface is usually poor, meaning that achieving large transparency modulation over large areas (> 0.5 cm) could be demanding.

Alternative reports on THz graphene saturable absorbers include multi-layer graphene grown on the carbon-face of silicon carbide, allowing a maximum absorption modulation of ~10%, inherently limited by disorder [10], or multilayer graphene films, grown *via* CVD on Nickel, in which a 10% transparency modulation, dominated by intraband phenomena has been shown [7].

D) SWITCHES FOR ELECTRONIC WAVES

When light is focused onto a nanometer-sharp metallic tip, miniature waves propagate on the surface of the underneath material in a circular fashion, starting from the tip apex. Such miniature waves can have the potential to be used in future compact electronic devices for lightning-fast information transport. However, to do that, these waves have to be ideally switched on and off at ultrafast timescales.

Recently, SiO_2/black phosphorus/SiO_2 heterostructures have been innovatively employed to this purpose [11]. Upon irradiation of a sequence of thin flakes of SiO_2/black phosphorus/SiO_2 by intense light pulses, freely moving electrons are generated inside the material. Without these electrons, no surface waves are present and the structure is switched "off". However, as soon as the first laser pulse generates the free electrons, a subsequent pulse can start the propagation of surface plasmons from the tip. The expansion of the plasmon waves can be traced in slow motion snapshots, unveiling *fs* switching times [11].

CONCLUSIONS

In conclusion, the superior mechanical pliability and the exceptional optical and transport properties of 2D materials and vdW heterostructures enclose an enormous potential for developing a novel generation of devices, optical components and systems in the THz frequency range (1–10 THz). Exceptional implications can be envisaged in frontier research fields as signal processing and computer technologies, ultrafast optics, plasmonics, and quantum metrology. Large area material production combined with large mobility, are the critical ingredients for a stable and reproducible production of the above technologies, needed for a progressively broader impact on market and research.

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