Graphene-oriented research has had a dramatic impact during the last decade. The superior carrier mobility, induced by the massless Dirac fermions, combines in graphene with a gapless spectrum that, although beneficial for applications requiring frequency-independent absorption, also prevents the effective switching of its conductivity in electronic devices and the related achievement of high on/off current ratios. Finite and direct band gaps are desirable for a wealth of applications, including transparent optoelectronics, photovoltaics, and photodetection. As an example, the performance of graphene detectors in the visible is strongly limited by the large dark currents that dominate under non-zero bias operation.

These issues are driving present research in the quest for alternative 2D materials: as a prototypical example, single-unit-cell thick layers of transition-metal dichalcogenides (TMDCs: MoS₂, MoSe₂, WS₂, WSe₂, etc.) have recently emerged as a valuable alternative. 2D TMDCs can be obtained from bulk crystals by employing the micromechanical exfoliation method, like for the case of graphene, but they show a direct band gap (0.4–2.3 eV), which enables applications that well complement graphene capabilities. In particular, 2D TMDCs are suitable for photovoltaic applications and for devising robust ultra-thin-body-field-effect transistor (FET) architectures which can easily provide subthreshold swings of \( \approx 60 \text{ mV}\text{ dec}^{-1} \) and \( I_{\text{on}}/I_{\text{off}} \) ratios up to \( 10^8 \). Nonetheless, their relatively low mobility (\( \leq 200 \text{ cm}^2\text{ V}^{-1}\text{ s}^{-1} \)) remains a major constraint for high-frequency electronic applications.

A good trade-off between graphene and TMDCs is represented by a novel class of atomically thin 2D elemental materials: silicene, germanene, and phosphorene. Among them, the latter one allows a peculiar single- or few-layer isolation from its bulk phase, e.g., black phosphorus (BP). Unlike silicon and germanium, BP, the most stable allotrope of the phosphorus element in standard conditions, shows a layered graphite-like structure, where atomic planes are held together by weak Van der Waals forces of attraction, thus allowing the application of standard micro-mechanical exfoliation techniques. As in graphene, each BP atom is connected to three neighbors, forming a stable layered honeycomb structure with an interlayer spacing of \( \approx 0.3 \text{ Å} \). In contrast with graphene, the hexagonally distributed phosphorus atoms are arranged in a puckered structure rather than in a planar one (Figure 1a). This property generates an intrinsic in-plane anisotropy that results in a peculiar angle-dependent conductivity, and an intrinsic optical anisotropy at visible and near-IR frequencies. Bulk BP has a small direct band gap of \( \approx 0.3 \text{ eV} \); the reduction of the flake thickness leads to quantum confinement which further enhances the gap up to \( E_g \approx 1.0 \text{ eV} \) in the limit case of phosphorene (a single layer of BP). As a result, the \( I_{\text{on}}/I_{\text{off}} \) ratio of a BP-based FET can be improved by employing thinner flakes: an \( I_{\text{on}}/I_{\text{off}} \) ratio of \( 10^5 \) has been recently reported in back-gated FET structures. On the other hand, thickness reduction is detrimental for carrier mobility: thinner flakes are more vulnerable to scattering by interface impurities and the effective mass of charge carriers increases when the number of atomic layers is reduced. Despite this, BP thin films are endowed with hole mobilities exceeding 650 cm² V⁻¹ s⁻¹ at room temperature (RT) and well above 1000 cm² V⁻¹ s⁻¹ at 120 K,[14] thus overtaking the limiting factor of large-gap TMDCs and allowing to reach high-frequency operation up to 20 GHz.[18] For all the reasons above, BP represents an ideal material for infrared optoelectronic applications and high-speed thin film electronics. Furthermore, the achievable superb \( I_{\text{on}}/I_{\text{off}} \) makes BP well suited for detection of terahertz (THz)-frequency light, being finite and direct band gaps, huge carrier density tunability, and large mobilities undisputed benefits in the viewpoint of low-dark current, large responsivity \( R_\nu \), and large modulation frequency (GHz). In addition, BP can be easily integrated with other photonic or optoelectronic components based on alternative 2D materials, like graphene, or with silicon technologies.

Photodetection relies on the conversion of absorbed photons into an electrical signal. This can be accomplished by several different mechanisms that have been recently reported in 2D...
FETs, like photothermoelectric, bolometric, or plasma-wave rectification effects. The device geometry, the inherent material thermal/electric properties, and the frequency of impinging light determine the dominant detection process.

To date, light detection at THz frequency in nanoscale FETs has been demonstrated to occur via the rectification of plasma waves in the transistor channel, induced by the external ac electric field. When an electromagnetic beam is coupled between the source (S) and the gate (G) electrodes, it excites carrier density oscillations, which, in turn, generate a driving longitudinal electric field through the channel. This simultaneous modulation of carrier density and drift velocity results in the onset of a dc signal that can be measured at the drain (D) electrode as a voltage, if D is kept in an open circuit configuration (photovoltage mode), or as a current, if D is in a short-circuit line profile, acquired along the dashed green line, is shown on the graph.

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FETs allow for the integration, as active channels, of a variety of 1D[21,24] or 2D[6,23] structures whose properties can either be exploited for[21,24] or investigated via[25] THz detection experiments. Conceiving and exploiting new material combinations can open the path to ground-breaking implementations of active devices and passive components across the intriguing and underexploited THz frequency range.

Here, we demonstrate efficient RT THz nanodetectors combining top-gated FETs exploiting thin BP flakes with integrated THz asymmetric antennas designed to enhance the sensitivity.

BP crystals were grown via chemical vapor transport techniques. Flakes having thickness ≤10 nm were then mechanically exfoliated from bulk BP crystal using a standard adhesive tape technique on a 300 nm thick SiO_2 layer on the top of a 300 μm intrinsic silicon wafer. The thickness of individual BP flakes was assessed via a combination of optical microscope mapping (Figure 1b), scanning electron microscopy imaging (Figure 1c), and atomic force microscopy (AFM) imaging (Figure 1e). Reproducible correlation has been found between the color map of the flake, as seen under the optical microscope (Figure 1b), and its thickness measured via AFM (Figure 1d).

Micro-Raman spectroscopy experiments with linearly polarized, z-incident excitation have been performed to evaluate the crystalline quality of BP flakes as well as to define the flake crystallographic orientation. The Raman spectra present three characteristic peaks at 362, 440, and 468 cm⁻¹ (Figure 1d) corresponding to the A_g⁰, B_2g, and A_g² vibrational modes observed in bulk BP, whose relative intensities change if the incident light is polarized along the armchair (x) axis, the zigzag (y) axis, or in-between, at a 45° angle. The topographic AFM scan of a single transferred flake (Figure 1d) shows a layered structure with two visible stacks with thicknesses 6.2 nm and 1.7 nm, respectively. By measuring a set of similar flakes we found that every stack has a thickness corresponding to an integer multiple of ~0.61 nm, i.e., the thickness of a BP monolayer (phosphorene).

To devise the FET detectors, we selected flakes with thickness h = 10 nm, roughly coincident with the out-of-plane screening length[27] and corresponding to ~16 layers of phosphorene. This choice ensures an ideal trade-off condition between high mobility and large carrier density tunability.[14] THz detectors have been realized by exploiting a combination of electron beam lithography (EBL) and metal evaporation (see the Experimental Section). The S and G electrodes were patterned in the shape of two halves of a planar bow-tie antenna (Figure 2c) having a total length 2L = 500 μm and a flare angle of 90°, in resonance with the 0.3 THz radiation. Figure 2a-c shows the device layout: the channel length (source-to-drain distance) has been set to L_c = 2.78 μm, the gate length is L_g = 580 nm, the average channel width is W = 2.6 μm, and the S–D contacts...
have been oriented along the armchair (x) BP axis (as revealed by the micro-Raman map), which provides the highest electrical conductivity and the lowest in-plane lattice thermal conductivity at RT.\cite{29} Under this configuration and in the presence of an 80 nm thick oxide layer, we simulated the geometrical gate-to-channel capacitance ($C_{gc}$) with a commercial 3D-FEM simulation software (COMSOL Multiphysics) and found $C_{gc} = 1.1 \, \text{fF}$.

The RT transport characterization was carried out using two dc voltage generators to drive the source-to-drain voltage ($V_{SD}$) and the gate voltage ($V_G$) independently (Figure 3). The device shows an ohmic behavior, with no signature of a contact Schottky barrier at RT. The transconductance characteristics have been acquired while sweeping $V_G$ in a limited bias range (see Figure 3) to prevent the top-gate insulating layer from breakdown. The resulting device is a p-type depletion mode FET with a visible hysteresis connected with the direction of the gate sweep. This effect is likely due to charging trapping in the top oxide layer and at the BP–dielectric interfaces.\cite{29} The ambipolar transport, typical of BP-based FETs,\cite{34,30} has not been observed in the present case due to the use of Cr/Au contacts that can significantly alter the band alignment and eventually suppress the electron conduction even at high positive $V_G$.\cite{31}

The achieved maximum transconductance $g_{mn} = 150 \, \text{nA V}^{-1}$ is independent of the gate sweep direction (dashed-dotted line in Figure 3a) and the $I_{on}/I_{off}$ ratio $= \times 10^{3}$, provides the needed carrier density tunability behavior.

In a 2D FET the mobility is conventionally extrapolated in a back-gate configuration\cite{10,30} or via capacitance-to-$G$ ($C–G$) measurements.\cite{29} In the present configuration, these strategies are difficult to be implemented because: (i) back gates require doped substrates, which are detrimental for THz detection and (ii) $C–G$ measurements are largely affected by the huge shunt capacitance induced by the presence of the bow-tie antenna. Here, we have adopted a different method to extrapolate the field effect mobility ($\mu_{FE}$) indirectly from the transconductance $g_{mn}$ curve, through the relation $g_{mn} = \mu_{FE} C_{ox} V_{SD} W/L_G$, where $C_{ox} = C_{TG}/A_C$ is the oxide capacitance per unit area, $C_{TG}$ is the top-gate capacitance and $A_C$ the gated area. This relation holds if the mobility is constant over the gate length and does not depend on the applied bias, as it is reasonable to assume in a top-gated system. For a thin layer channel, $C_{TG}$ (Figure 3b, inset) is given by the sum in series of the geometrical capacitance $C_{G}$ (whose value can be obtained via FEM simulations) and the parallel of the interface trap capacitance ($C_q$) and the quantum capacitance ($C_G$):\cite{29,32}

$$C_{TG} = \left( \frac{1}{C_{G}} + \frac{1}{C_{q}} \right)^{-1}$$

The quantum capacitance is related to the material density of states and takes into account the fraction of $V_G$ dropped within the channel to modify the carrier population. Therefore, according to Equation (1), the gate voltage modulates the band structure (via $C_{G}$) and simultaneously fills trap states (via $C_{q}$) and carrier states (via $C_{G}$). An estimation of the quantity ($C_{q} + C_{G}$) can be obtained from the subthreshold swing ($S_{th}$) of the FET. The logarithmic plot of the transconductance curve shows a linear region (shaded areas in Figure 3b) whose slope corresponds to the subthreshold slope ($S_{th}$)\cite{1}: this regime identifies the onset of a pure hole thermionic emission current over the potential barrier generated at positive $V_G$. In general, $S_{th} = 60 \, \text{mV dec}^{-1}$, where $\beta$ is the band movement factor: $\beta = [1 + (C_{q} + C_{G})/C_{G}]$. In the subthreshold regime, the channel is almost depleted of free carriers, hence $C_{q}$ can be safely neglected: the channel bands move one-to-one with the applied gate bias.\cite{31} In the present case, we estimate $\beta = 7$ (independently from the $V_G$ sweep direction) from the linear fit to the data (shaded areas, Figure 3b), then $C_{q} = 6 \, C_{G}$; by varying $V_G$, the top-gate capacitance will vary in the range $[6/7 \, C_{G} ; 6 \times C_{G}]$.

From these considerations, we can infer a lower bound for $\mu_{FE}$:
\[ \mu_{\text{FE}}(\text{TG}) = \frac{g_m \cdot I_c}{C_{\text{pe}} V_G} \]  

(2)

corresponding to \( \mu_{\text{FE}} = 470 \, \text{cm}^2 \, \text{V}^{-1} \, \text{s}^{-1} \).

In order to prove THz detection at RT, we employed an electronic source whose output frequency was tuned in the range 0.26–0.38 THz. The output beam, having polarization parallel to the antenna axis, was collimated by a set of two 1/f off-axis parabolic mirrors in a 4 mm diameter spot while its amplitude was modulated as a square wave at 618 Hz by means of a mechanical chopper. The photoresponse was measured in photovoltage mode: with the S electrode grounded, the response voltage \( \Delta u \) was collected at the D electrode in an open circuit configuration (see the Experimental Section).

Figure 4a shows the channel conductance dependence on the gate bias, measured while the 0.298 GHz line of a tunable source (see the Experimental Section) is impinging on the device. The optimal frequency has been preventively selected after testing the frequency dependence of the measured photovoltage (Figure 4b), that is inherently set by the antenna design.

Under our experimental regime, the THz-induced plasma waves propagating in the transistor channel are expected to be strongly damped, being \( 2\pi \nu \tau = 1 \). Here, \( \tau = m_e^* \nu / e \) is the hole relaxation time (\( \leq 200 \, \text{fs} \)) and \( m_e^* \) is the hole effective mass along the armchair direction.\(^{[17]}\)

In the physical regime in which the detection mechanism is mediated by the rectification operated by the self-mixing of overdamped charge density waves, the transfer characteristics allow predicting the expected generated photovoltage,\(^{[6,21–24]}\) which is proportional to:

\[ \Delta u_T \propto -\frac{1}{\sigma} \frac{d \sigma}{dV_G} \left[ \frac{R_i}{\sigma + R_i} \right] \]  

(3)

where the minus sign accounts for the hole majority carriers, and in which loading effects arising from the finite impedance \( R_i \) of the measurement setup have been properly included.

Conversely, if the THz light induces thermoelectric effects, the photothermoelectric voltage is proportional to the Seebeck coefficient \( S \) that is related to \( \sigma \) via the Mott equation: \(^{[6]}\)

\[ S = -\frac{\pi^2 k_B T}{3e} \left( \frac{1}{\sigma} \frac{d \sigma}{dV_G} \right) dE_f \]  

(4)

where \( k_B \) is the Boltzmann constant, \( e \) is the electron charge, and \( E_f \) is the Fermi energy (see the Experimental Section).

Figure 4c shows the predicted \( \Delta u_T \) trend as a function of \( V_G \): the THz response is expected to peak around \( V_G = +3.5 \, \text{V} \). The comparison with the experimental responsivity/photovoltage curve (Figure 4e) reveals a good qualitative agreement, unveiling that the rectification of THz-induced overdamped plasma waves, in the BP channel, might be the dominant detection process. However, it is worth noticing that, at negative gate biases, \( R_c \) does not decrease to zero (Figure 4e), in clear contrast with the model predictions, but saturates at an average value of \( \approx 0.03 \, \text{V} \, \text{W}^{-1} \). This suggests a contribution to the photodetection process. Such a conclusion is well supported by the fact that being the material thermal conductance anisotropic and since the current is flowing along the armchair (low thermal conductance) axis, our geometry significantly enhances BP thermoelectric performance.\(^{[28]}\)
A maximum responsivity \( R_v = 0.15 \) V W\(^{-1}\) has been reached, slightly higher than that achieved in bilayer graphene FETs fabricated under identical geometries. Very differently from graphene bilayer FET,[23] the responsivity curve is clearly unaffected by interband transition–related intrinsic noise features. For sake of comparison the measured net residual noise, estimated by measuring the photocurrent signal while obscuring the THz beam with an absorber, is shown in Figure 4e (black curve).

The noise-equivalent power (NEP) has been extracted from the ratio \( N_{\text{th}}/R_v \), by assuming that the main contribution to the noise figure is the thermal Johnson–Nyquist noise \( (N_{\text{th}})\),[23] associated with the non-zero resistance of the FET channel. Although this hypothesis neglects the 1/f and the shot noise contributions, it provides a lower limit for the NEP.[21–23] Minimum values of \( \approx 40 \) nW Hz\(^{-1}\) have been reached, comparable with the NEP of the best bilayer graphene-based THz photodetectors, operating at similar frequencies, tested under identical antena-coupling configuration and exploiting comparable geometries. The achieved results highlight the potential of BP for devising novel active THz devices.[23] Major improvements can be envisioned by engineering wide gate architectures and impedance matched nano-antennas. Furthermore, by exploring the inherent in-plane material anisotropy, BP appealingly opens the perspective to devise alternative device geometries that could allow properly “engineering” the preferred photodetection mechanism to address targeted terahertz application requirements.

Experimental Section

Fabrication: Micro-Raman spectroscopy on the exfoliated BP flakes was performed with a Renishaw (InVia) system, equipped with a frequency doubled Nd:YAG laser, exploiting the 532 nm line and having maximum output power of 500 mW (CW). In the present experiments, the optical intensity was kept at 0.04 mW \( \mu \)m\(^{-2}\), since intensity values larger than 0.8 mW \( \mu \)m\(^{-2}\) are likely to damage the flakes. The S–D FET channel was defined via a combination of EBL and thermal evaporation to deposit the (10/70 nm) (Cr/Au) S and D metal contacts. An 80 nm thick SiO\(_2\) oxide layer was then deposited on the sample via Ar sputtering. The G electrode was patterned on the oxide via EBL and a (10/90 nm) (Cr/Au) layer thermally evaporated on it. Although high work-function metals, like Pd or Ni, are preferable to achieve ohmic contacts on p-type semiconductors,[30] chromium ensures better adhesion to the SiO\(_2\) substrate, which turned out to be crucial to safely lift-off large-area planar metallic structures. Furthermore, the electrical performances of BP-based FETs are prone to time degradation due to ambient air humidity.[33] therefore surface passivation strategies have to be adopted. The deposition of an insulating oxide layer over the exposed face of a BP-flake proved to be reasonably effective.[34] In this case, FETs were indeed electrically characterized after three weeks of exposure to the environment, finding a 15% degradation in the channel conductance with respect to postprocessing tests. Storing the devices under vacuum, about 3% degradation in the conductivity and on/off current ratios over two months has been achieved. Further improvements can be envisaged if SiO\(_2\) is replaced by high-k dielectrics (Al\(_2\)O\(_3\), HfO\(_2\), etc.), which are expected to provide more efficient protections.[34]

Optical Testing: The voltage response \( (\Delta u) \) was recorded at the D electrode in an open circuit configuration, while keeping the S electrode grounded, by means of a lock-in amplifier, connected to a low-noise voltage preamplifier having an input impedance of 10 MQ and a gain factor \( G_u = 1000 \). \( \Delta u \) was retrieved from the voltage signal read on the lock-in (LIA) via the relation \( \Delta u = 2.2 \text{LIA}/G_u \), where the factor 2.2 accounts for the square wave modulation, since the lock-in measures the rms of the sine wave Fourier component. The highest photocurrent was achieved at 0.298 THz; the corresponding source power, measured at the focal point of the optical system, was 325 \( \mu \)W. The detector responsivity \( (R_v) \) was then extracted from \( \Delta u \) through the relation \( R_v = (\Delta u - S_I)/((P_{\text{beam}} - S_I)) \), where \( S_I \) is the beam spot area (\( S_I = \pi d^2/4 \), where \( d \) is the spot diameter) and \( S_I \) is the active detection area. In the present geometry, the area of the 500 \( \mu \)m long bow-tie antenna is smaller than the diffraction limited one (\( S_I \)), hence we assume \( S_I = S_A = \pi d^2/4 \).

Thermoelectric Effect: Equation (4) has been derived starting from the BP bidimensional carrier density equation \( n(E_f)\),[27] via the relation:

\[
\left( \frac{dn}{dE_f} \right) = \frac{\partial \sigma}{\partial E_f} \left( \frac{d\sigma}{dV} \right) \frac{dV}{dE_f}
\]

where \( h \) is the flake thickness. This equation holds in the range in which the mobility is independent from carrier energy. In the present case, this is well supported by the limited gate voltage operation range and by the femto-Farad order \( C_{\text{gb}} \) capacitance value. Exploiting the relation \( dV/dE_f = \beta \), that occurs in the subthreshold region, where the quantum capacitance \( C_q \) is negligible with respect to \( C_{\text{gb}} \) and \( C_v \), it is possible to correctly overlap the calculated \( dn/dE_f \) and the measured \( d\sigma/dV \) to extract the term \( dV_C/dE_f \) over the whole gate voltage range.

Acknowledgements

This work was partially supported by the Italian Ministry of Education, University, and Research (MIUR) through the program FIRB—Future in Ricerca 2010 RBFR10UL5P “Fundamental research on Terahertz photonic devices” and by the European Union through the MPNS COST Action “MP1204 TERA-MIR Radiation: Materials, Generation, Detection and Applications.” W.K. acknowledges the National Science Poland Centre (DEC-2013/10/M/ST3/00705). J.H. was supported by NSF/ LA-SiCMa program under Award No. EPS-1003897. A.P. thanks Anna Cupoliillo for helpful discussions.

Received: April 29, 2015
Revised: July 8, 2015
Published online: August 13, 2015

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