Physica E 85 (2017) 164–168

Contents lists available at ScienceDirect

Physica E

journal homepage: www.elsevier.com/locate/physe

Plasmonic ambient light sensing with MoS_2-graphene heterostructures

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HIGHLIGHTS

- Plasmonic photodetection of MoS_2-graphene heterostructures.
- Hot-carrier plasmonic doping of MoS_2 mechanism behind this photodetector.
- Responsivity is in agreement with photopic standards of the luminosity function.
- The response time of the detector is less than the eye blinking time, i.e 0.1 s.

ARTICLE INFO

Article history:
Received 9 June 2016
Received in revised form
23 July 2016
Accepted 27 August 2016
Available online 29 August 2016

ABSTRACT

We present experimental results on plasmonic photodetection of ambient light using MoS_2-graphene heterostructures illuminated with three very-low-power light emitting diodes (LEDs) radiating in blue, green, and red, respectively. The working principle of this photodetector validates the recent predictions of hot-carrier plasmonic doping of MoS_2. The obtained responsivity for each spectral domain is in agreement with photopic standards of the luminosity function. The response time of the detector is less than the eye blinking time.

1. Introduction

Sensing ambient light is deeply involved in our daily life. It can be encountered, for example, in automatic brightness control, automatic turn-off in any touch-screen display, including that of mobile phones, or automatic activation of keypad lightning and screen brightness adjustments in laptops. The application list is very long, involving almost all consumer electronics products such as digital cameras, television, printers, games and automotive applications. A good survey of photonic sensors is found in [1], while the applications of sensing ambient light are detailed in [2].

The target of any ambient light detector, that of having a spectral response close to the human eye, prompted the International Commission of Illumination (CIE) to establish a standard photopic luminosity function, which imposes certain responsivity values (in A/W) on the entire visible spectrum from 400 nm to 700 nm, in particular for blue, green and red light. The main problem is that nowadays ambient light detection is based on Si devices, which show significant mismatch with the standard curve of CIE luminosities and human eye because the optical response spectrum of Si detectors extends to IR due to the small energy bandgap of this material (1.11 eV). The fulfilment of the CIE standard was achieved up to now only with AlGaAs ambient light detectors [3] in a complicated configuration containing 11 vertical heterostructures between the substrate and the metallic contacts.

Molybdenum disulfide (MoS_2)-graphene heterostructures were proposed as an alternative ambient light detector, because the spectral response of mono- and few-layers MoS_2 is located mainly in the visible region [4]. Indeed, the bandgap of bidimensional (2D) MoS_2 is higher than that of Si, monolayer MoS_2 being a direct semiconductor, with a bandgap of about 1.9 eV, the bandgap decreasing as the number of layers increases until it reaches the value of 1.2 eV corresponding to bulk MoS_2 [5–8]. As such, in an ambient light detector MoS_2 could assure the absorption and thus the generation of charge carriers in a heterostructure comprising also a graphene layer, which mainly enhances the conduction properties of the heterostructure [9].

The state-of-the-art of optical applications of atomically thin MoS_2 monolayers and few-layers MoS_2, as well as of other 2D materials, can be characterized as a race for higher responsivities, i.e. higher quantum efficiencies. For instance, extraordinary photoresponses, of 4×10^2 A/W, were obtained in 2D In_2Se_3 nanosheets [10] deposited on interdigitated electrodes and illuminated at 2–4 W/m^2 in the spectral range of 300–500 nm, the area

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http://dx.doi.org/10.1016/j.physe.2016.08.026
1386-9477/© 2016 Published by Elsevier B.V.

of the device being of only 0.108 μm^2. Regarding the heterostructure of interest in this paper, ultrahigh-gain photodetectors based on atomically thin graphene-MoS_2 heterostructures in a field-effect-transistor (FET) configuration were demonstrated. These photodetectors, with an area of 2×10^{-4} cm^2 [9], show a responsivity of 10^6 A/W when illuminated with a continuous laser at 532 nm with a power density of 35.21 W/cm^2. In addition, gate-modulated photoresponse in a vertical graphene-MoS_2 heterostructure attaining internal quantum efficiencies up to 85% was demonstrated under illumination with a 80-μW-power laser at 514 nm, focused in a spot of 1 μm [11].

The goal of this paper is to investigate the photoresponse of a MoS_2-graphene heterostructure, in which the increase of charge carriers is not due mainly to absorption of ambient light but to injection of hot holes from Au into MoS_2 as a result of plasmonic generation of hot carriers in Au. As such, the proposed MoS_2-graphene photodetector validates the recent theoretical prediction of hot-carrier generation due to surface plasmon decay [12]. The MoS_2-graphene photodetector that we propose has a larger area than previous devices, being thus suitable for display applications, and responds even at weak optical powers, its spectral response matching that of the human-eye and thus fitting the CIE requirements. Such a device, which can be easily assembled, would confer simplicity of ambient light detection compared to standard semiconductor heterostructure technology, and less mismatches with CIE prerequisites than Si photodetectors.

2. Fabrication of the plasmonic MoS_2-graphene photodetector and experimental setup

Molybdenum disulfide pristine flakes in water/ethanol mixture, and 10 mm × 10 mm graphene on a nickel/SiO_2/Si substrate were supplied by Graphene Supermarket. Au deposition on glass was achieved through cathodic sputtering (50 nm Au with a 5 nm Cr adhesion layer). The gold sputtered glass was then immersed into the MoS_2 mixture, slowly retracted by hand, and allowed to dry out in open air. The graphene on nickel/SiO_2/Si substrate and the molybdenum disulfide decorated Au/glass were finally sandwiched together. A schematic device cross-section is shown in Fig. 1; the available transparent area is 0.5 cm^2. Graphene, grown by CVD [13,14] on Ni (thickness 400 nm) is not uniform, but forms patches, the entire film looking like a patchwork (see Fig. 2(a)). The thickness of the patches varies between one and few graphene monolayers, with an average of 4 monolayers. In turn, the MoS_2 layer deposited over glass/Au is non-uniform also, the resulting grainy structure being visible in Fig. 2(b).

In contrast to previous photodetectors based on MoS_2-graphene heterostructures, where the incoming light is mostly absorbed in the MoS_2 layer, which has a much higher absorption coefficient than graphene, and the produced photocarriers are separated by the internal field associated to the Schottky barrier at the MoS_2-graphene interface, our photodetector is based on a different physical principle. Indeed, illumination through the metallic Au layer precludes the transmission of a significant fraction of the incident electromagnetic radiation. However, a significant photoresponse of the heterostructure is still possible due to plasmonic generation of hot carriers [12] in Au, followed by MoS_2 doping with hot holes, favoured by the fact that at ambient conditions, at the MoS_2-graphene interface, electrons move from MoS_2 to graphene [9]. Due to the high number of trapping centers in MoS_2 flakes, the injected holes are trapped and act as an

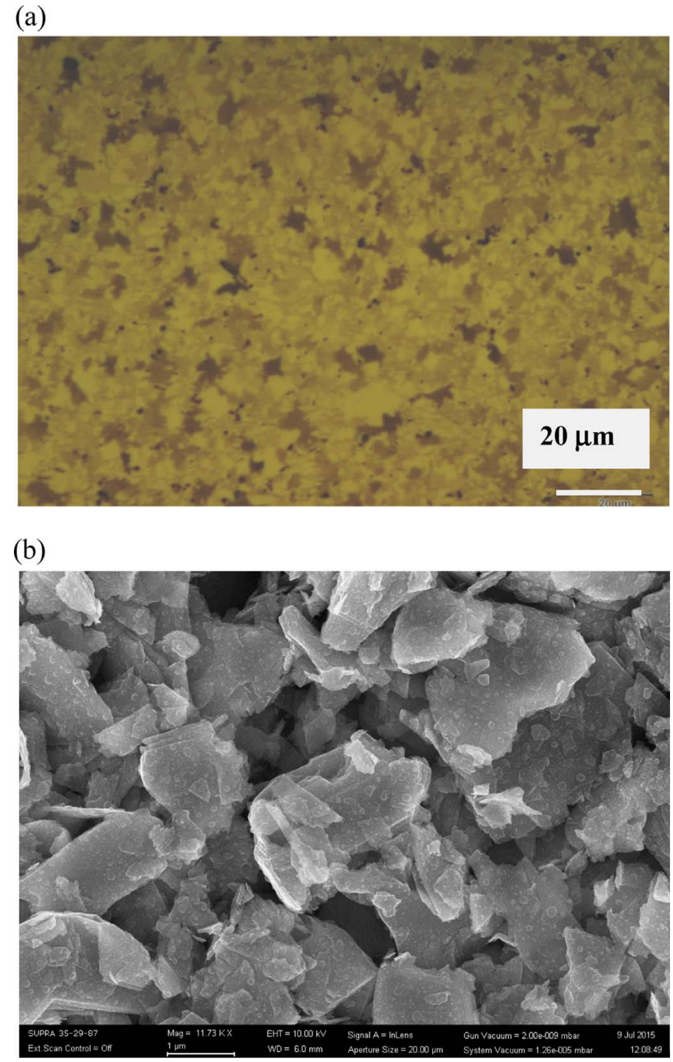

Fig. 2. (a) Optical image of graphene/Ni, (b) SEM photo of MoS_2 deposited on Au.

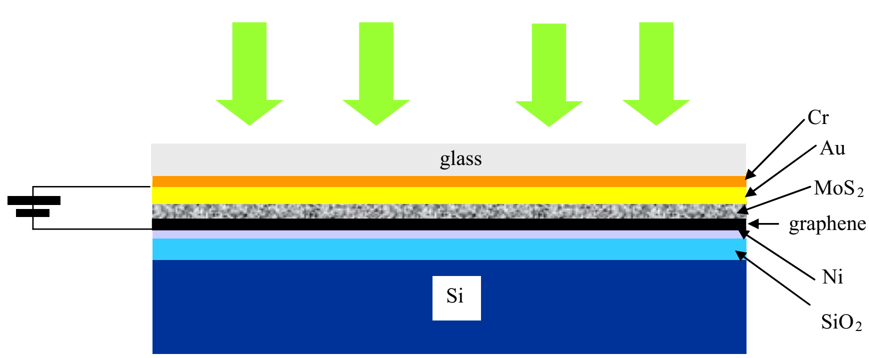

Fig. 1. Schematic representation of the MoS_2-graphene heterostructure.

effective applied positive gate voltage for graphene, shifting the Fermi energy level in the latter material. The photo-doped MoS_2 enhances therefore the photocurrent by increasing the carrier concentration in graphene. Experimental evidence of plasmonic hot carrier generation at MoS_2-metal interfaces [15], leading in particular to an enhanced photoresponse [16], have been obtained recently.

Despite the extreme simplicity of the fabrication of the MoS_2-graphene heterostructure presented above, the nonuniformities in the MoS_2 layer could be seen as a disadvantage. However, when we tried to increase the MoS_2 uniformity by increasing the thickness of MoS_2, the photoresponse vanished. This is most probably due to a significant decrease in the efficiency of charge carrier collection caused by an enhancement of trapping/recombination of charge carriers in MoS_2.

LED sources in combination with a LED Driver (MetrohmAutolab B.V.) were directed on the MoS_2-graphene heterostructure at normal incidence through the glass substrate, and the electrical response at various optical excitations was obtained and processed with the help of the Autolab/PGSTAT302N electrochemical system from Eco Chemie. The blue (470 nm), green (530 nm), and red (627 nm) LED sources used in our experiments were calibrated before the measurements. Light source calibration was performed using a calibrated photodiode (model FDS100 – CAL, range 350–1100 nm) from Thorlabs.

3. Results and discussions

The plasmonic MoS_2-graphene heterostructure photodetector was exposed to the light source (blue, green or red LEDs) at a controlled distance (20 cm) on the optical bench. The calibration was performed using the Autolab/PGSTAT302N electrochemical system in combination with the LED Driver, which controlled the driving current applied to the LEDs.

We have displayed in Fig. 3 the measured currents as a function of the applied voltages at various optical power densities, when the device is illuminated with the blue LED. The reproducibility of the results was validated using a couple of structures and making repetitive measurements in single and dual sweep mode. In Fig. 3, the current-voltage dependence at dark and at illumination with the blue LED at two optical densities were represented in two situations: (i) as measured, and (b) in the logarithmic scale. The same representations for the case of the red and green LED light sources are illustrated in Figs. 4(a)-(b) and 5(a)-(b), respectively.

In Fig. 6 we have represented the current of the heterostructure at ON-OFF illumination at different optical power densities for the green LED source; similar dependences have been observed for the blue and red LEDs. The response time of the photodetectors is 0.1 s in all cases, value that is smaller than the eye blinking time, which is about 0.3–0.4 s

From Figs. 3–5 we can see that we have an almost symmetric ambipolar Schottky device behaviour, characterized by strong current nonlinearity, although the device structure is asymmetric, the MoS_2-graphene heterostructure being sandwiched between Ni on one side and Au on the other. As the current is mainly due to graphene, the Fermi level of which is modified by the effective gating of photo-doped MoS_2, the symmetry of the *I-V* characteristic must reflect the symmetry of the Schottky barriers at the two polarizations. Whether it is difficult to estimate the workfunction at the graphene/Ni, MoS_2-graphene and Au-MoS_2 interfaces, because of the extreme sensibility of this parameter on the quality/charge impurities of these interfaces involving atomically-thin materials [9] and the number of layers of the MoS_2 flakes, experimental data show that the effective Schottky barriers for the two polarizations are almost identical. This result is in agreement

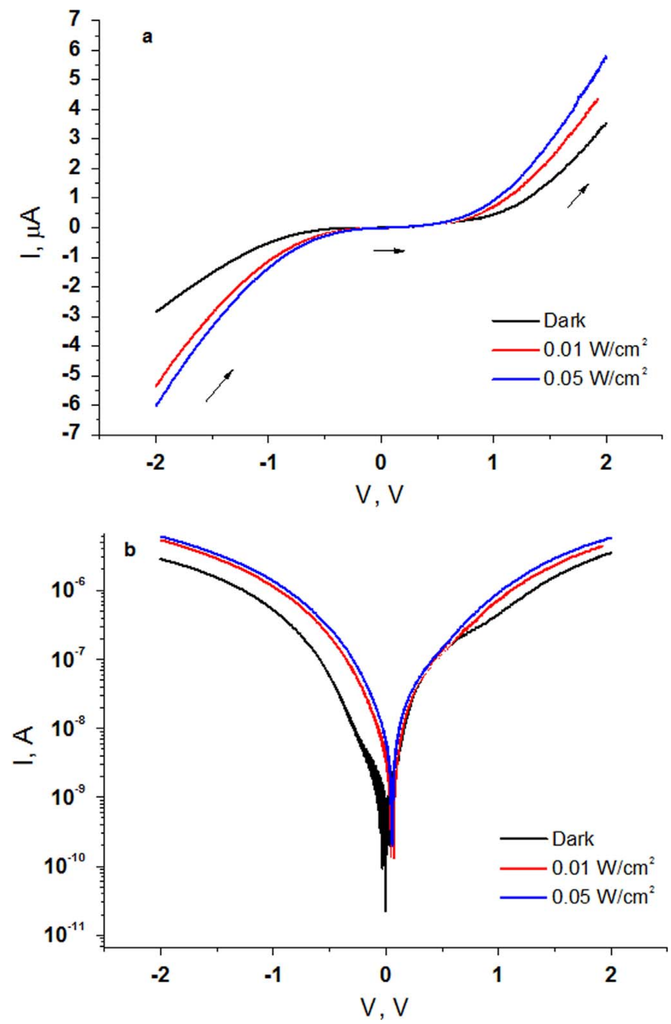

Fig. 3. The current-voltage dependence in the case of blue LED illumination: (a) as measured, (b) logarithmic scale.

with the band alignment between graphene on one hand, and *p*-MoS_2, Au, and Ni on the other hand, as revealed in Ref. [17], for example.

In order to characterize the device, we have calculated first the ideality factor of the MoS_2-graphene heterostructure in the dark state, and have found a value of 1.8. The ideality factor decreases when the structure is illuminated, and becomes 1.15 at an optical power density of 0.06 W/cm^2 for illumination with the red LED, 1.07 for illumination with the green LED at an optical power density of 0.018 W/cm^2, and nearly 1 for blue LED illumination at an optical power density of 0.05 W/cm^2. The ideality factor of 1.8 at dark is consistent with previous studies of exfoliated graphene-MoS_2-metal vertical heterostructures, which showed that this parameter depends on the number of MoS_2 layers [18], taking values higher than 1 for a number of MoS_2 layers smaller than 15.

In general, ideality factors close to 2 occur when recombination of carriers takes place in the space charge region, mediated especially by recombination centers resulting from interface or mid-gap states, whereas an ideality factor of 1 characterizes an ideal junction, in which band-to-band recombination of charge carriers occurs only in the neutral regions. A value of 1.8 for the ideality factor of the MoS_2-graphene heterostructure at dark suggests a high concentration of defects/recombination centers. These trapping centers are progressively filled by plasmonic photogenerated hot carriers as the incident power increases, as supported by our finding that the ideality factor decreases at

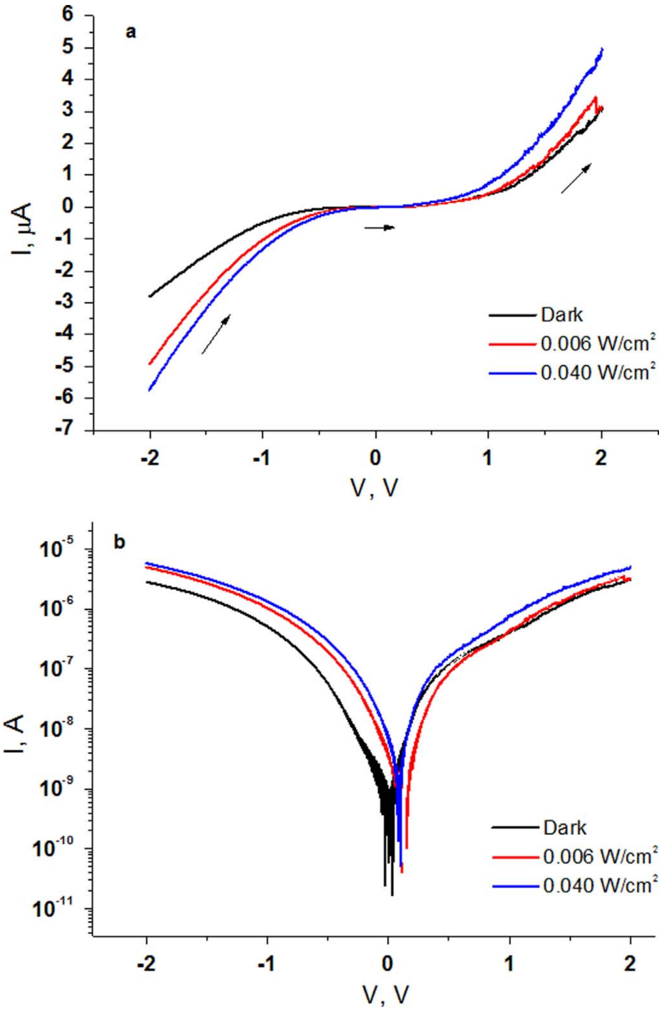

Fig. 4. The current-voltage dependence in the case of green LED illumination: (a) as measured, (b) logarithmic scale.

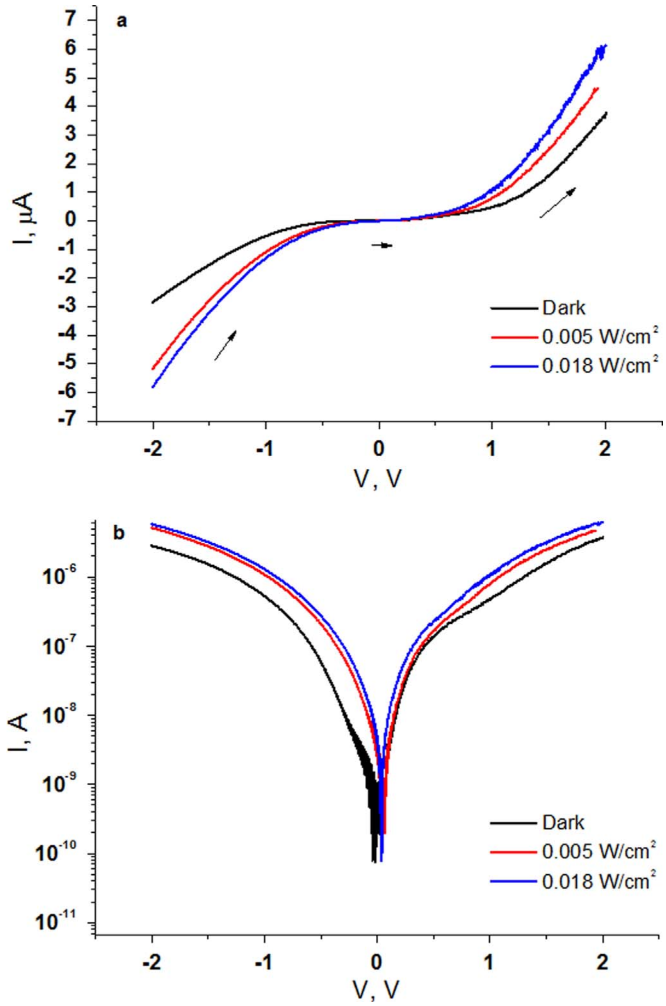

Fig. 5. The current-voltage dependence in the case of red LED illumination: (a) as measured, (b) logarithmic scale.

illumination. Moreover, the progressive filling of trapping centers as the number of photocarriers increases is also suggested by the shift of the voltage at which the current is minimum, observed in Figs. 3(b), 4(b) and 5(b). In all our experiments the voltage for which the current is minimum shifts initially to higher values at weak illumination (consistent with an effective positive gating of graphene), and then to lower values (but still higher then at dark) at stronger illumination. The shift of this voltage value at illumination is a consequence of the competition between two phenomena: a shift of the effective Fermi level in graphene as a result of effective gating of photo-doped MoS_2 [9], and the screening of the electrostatic potential in the structure due to an increased charge trapping in MoS_2. The second mechanism, inducing a shift to lower values of the voltage for which the current is minimum, becomes more significant for higher illumination powers.

The responsivities for the three LED sources were calculated at an applied voltage of 2 V and compared with CIE standards. For the case of green LED illumination, we have obtained $R=83$ mA/W at an optical power density of 0.005 W/cm^2, while the CIE standard requires 80 mA/W at the same optical power density. Further, for illumination with the red LED at an optical power density of 0.06 W/cm^2 we find $R=41.6$ mA/W, while the CIE standard demands 30.1 W/cm^2, and for the blue LED case we have $R=6.3$ mA/W at 0.01 W/cm^2, a value which is very close to the CIE standard, which requires 10 mA/W. From these values, it is apparent that the highest responsivity is obtained for green illumination, although

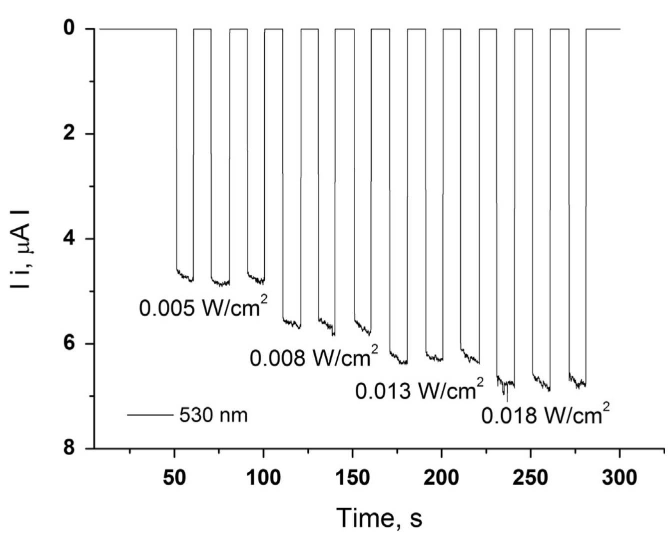

Fig. 6. Measured current at ON-OFF illumination at different optical power densities for the green LED source.

the optical power density is the smallest in this case. This fact supports the hypothesis of a plasmonic doping of MoS_2 with hot carriers, effect that is resonant in the green part of the visible spectrum.

4. Conclusions

We have analyzed a MoS_2-graphene heterostructure fabricated using a bottom-up approach. We have measured the current-voltage dependence of this device in the dark state and at very low optical power densities from calibrated blue, green and red LEDs. The ambipolar Schottky-like conduction is characterized by an ideality factor that is dramatically improved under illumination due to the progressive filling of the recombination centers in the MoS_2 layer with a grainy structure. The responsivities at 2 V at green and red LED excitations are above the CIE standards, while that at blue LED illumination is close to the CIE standard, making the device applicable for ambient light detection and sensing. The response time of the detector at ambient light variation is less than the eye blinking time. The main advantage of the plasmonic MoS_2-graphene heterostructure investigated in this paper as ambient light photodetector is its ease of fabrication.

Acknowledgements

This work was supported by project no. PN-II-RU-TE-2014-4-1095 of the Romanian National Authority for Scientific Research and Innovation, CNCS – UEFISCDI, and by the National Core R&D Programme – Nucleu.

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