A STUDY ON ULTRAVIOLET PHOTODETECTORS

and a Case Study on The Existing and Upcoming Technology of Graphene-based UV Photodetectors.

An M.Sc. Dissertation Report by:
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UV And UV Photodetection

Electromagnetic radiation comes from the sun and transmitted in waves or particles at different wavelengths and frequencies. This broad range of wavelengths is known as the electromagnetic (EM) spectrum. The spectrum is generally divided into seven regions in order of decreasing wavelength and increasing energy and frequency. The common designations are radio waves, microwaves, infrared (IR), visible light, ultraviolet (UV), X-rays and gamma-rays.
Ultraviolet (UV) light falls in the range of the EM spectrum between visible light and X-rays. It has frequencies of about 8×10^{14} to 3×10^{16} cycles per second, or hertz (Hz), and wavelengths of about 380nm to about 10nm.
It is produced by the sun, electric arcs and specialized lights, such as mercury-vapor lamps, tanning lamps, and black lights. Although long-wavelength ultraviolet is not considered an ionizing radiation because its photons lack the energy to ionize atoms, it can cause chemical reactions and causes many substances to glow or fluoresce. Consequently, the chemical and biological effects of UV are greater than simple heating effects, and many practical applications of UV radiation derive from its interactions with organic molecules.

(*UV Radiation | NCEH | CDC*, n.d.; *What Is Ultraviolet Light? | Live Science*, n.d.)

Introduction to UV detection

Photodetection in the ultraviolet (UV) region has drawn extensive attention owing to its various applications in industry, instrument, and our daily life. Ultraviolet research began in the latter half of the 19th century, when the invisible radiation beyond the blue end of the visible spectrum began to receive attention. It was soon realized that the Earth's atmosphere set limitations on ultraviolet research. For solar and celestial observations, stratospheric ozone limited the wavelengths reaching the surface of the Earth to about 300 nm. Most of the UV light from the Sun is absorbed by the atmospheric ozone layer. UV light is typically divided into four spectral regions.

near ultraviolet	NUV	400–300 nm
mid ultraviolet	MUV	300–200 nm
far ultraviolet	FUV	200–100 nm
extreme ultraviolet	EUV	100–10 nm

In addition to the above names, the following names for wavelength regions may be encountered:

vacuum ultraviolet	VUV	200–10 nm
deep ultraviolet	DUV	350–190 nm
ultraviolet-A	UV-A	400–320 nm
ultraviolet-B	UV-B	320–280 nm

Spectrographs carried to higher altitudes on mountains gave intriguing evidence that solar and stellar emissions continued to shorter wavelengths. In laboratory spectrographs, atmospheric molecular oxygen absorption limited the useful lower wavelengths to about 200 nm, unless the spectrograph could be placed in a vacuum chamber. The wavelengths shorter than 200 nm thus came to be called the vacuum ultraviolet. Because of the lack of good vacuum pumps and associated technology, research was difficult and not widely done. In addition to atmospheric limitations, optical methods used in the visible failed in the ultraviolet because of the lack of materials having good transmissivity and reflectivity.

In general, UV detectors fall into two categories: photon detectors (also named photodetectors) and thermal detectors. In photon detectors the incident photons are absorbed within the material by interaction with electrons. The observed electrical signal results from the changed electronic energy distribution. The photon detectors measure the rate of arrival of quanta and show a selective wavelength dependence of the response per unit incident radiation power. In thermal detectors, the incident radiation is absorbed and raises the temperature of the material. The output signal is observed as a change in some temperature-dependent property of the material. UV photon detectors (Fig. 1) have traditionally been devoted into two distinct classes, namely, photographic and photoelectric. Photographic emulsion has the great advantage of an image-storing capability and can thus record a large amount of data in a single exposure. However, photographic emulsion has a number of limitations: sensitivity is considerably lower than that of a photoelectric detector, the dynamic range is limited, the response is not a linear function of the incident photon flux at a specific wavelength, and

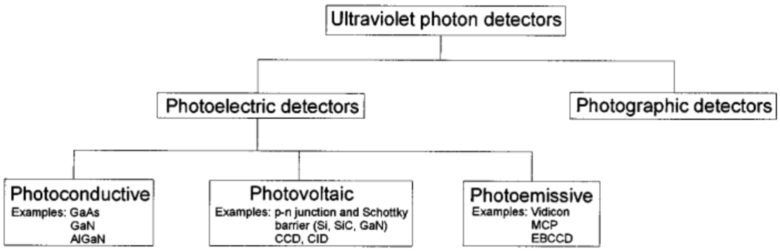


Figure 1
Classification of ultraviolet photon detectors.

emulsion is sensitive to a very wide energy range (accordingly the elimination of background fog levels induced by scattered light and by high-energy charged particles is extremely difficult). Photoelectric detectors, on the other hand, are more sensitive, have a greater stability of response and provide better linearity characteristics. In the last decade considerable progress in the image-recording capability of photoelectronic devices has been observed. Recently developed photovoltaic array detectors such as the charge coupled devices and photo-emissive array detectors for the first time combine the sensitivity and radiometric stability of a photomultiplier with a high-resolution imaging capability

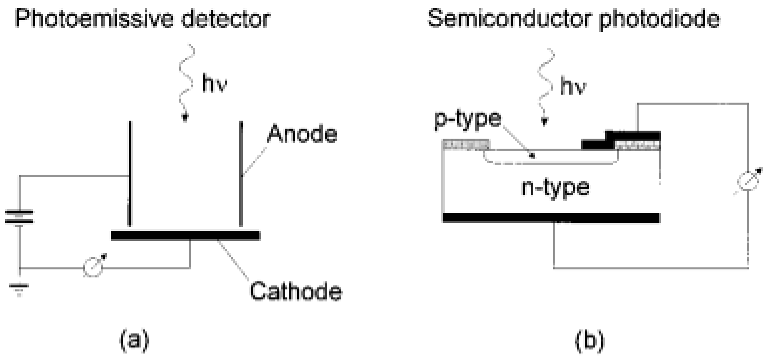


Figure 2
(a)Principal operation of photo-emissive and (b) semiconductor detectors

In the photo-emissive detectors, the primary photoelectron can be multiplied by the process of secondary emission to produce a large cloud of electrons. The occurrence of a single photoelectron event then can be detected either directly with conventional electronic circuits or by accelerating the electron cloud to high energy and allowing it to impact a phosphor screen. The emitted pulse of visible-light photons can then be viewed directly, or detected and recorded by additional photosensitive systems. Detectors operated in this pulse-counting mode can provide the ultimate level of sensitivity set by the quantum efficiency of the photocathode.

In the semiconductor detectors, the photons are absorbed in the bulk of the semiconductor material producing electron-hole pairs which are separated by an electrical field. These detectors make use of the internal photoelectric effect where the energy of the photons is large enough to raise the electrons into the conduction band of the semiconductor material.

Type	*Advantages*	*Disadvantages*
Photo-emissive detectors	• Easy to operate • High sensitivity • Solar blind	• Low quantum efficiency • Strong spectral dependence of responsivity • Sensitiveness to surface contaminations
Semiconductor detectors	• Broad spectral responsivity • Excellent linearity • High quantum efficiency • High dynamic range of operation • Large-format image arrays	• Induced aging effects

The importance of UV semiconductor detectors has resulted in the recent meteoric expansion of the semiconductor industry, and second, the continuing emphasis on the development of low-light-level imaging systems for military and civilian surveillance applications. These detectors should:

1. Not be sensitive to light at optical wavelengths (commonly referred to as being solar blind)
2. Have high quantum efficiency
3. Have a high dynamic range of operation
4. Have low backgrounds since noise arising from the background often dominates in faint UV observation.

A high-performance photodetector should satisfy the 5S requirements of *high sensitivity, high signal-to-noise ratio, high spectral selectivity, high speed, and high stability*.

(H. Chen et al., 2015; Donati, 2000; L.C. Passos & M.F.S. Saraiva, 2019; Razeghi & Rogalski, 1996)

Heterojunction UV detectors:

UV photodetectors that convert UV illumination into electric signals are fundamental optoelectrical components which have unique requirements for '5S' (sensitivity, selectivity, stability, signal-to-noise ratio, and response speed). To that end, it is a common strategy to construct heterojunctions for reducing carrier recombination, which can suppress the PD's photo-response to high-energy photons. Generally, heterojunctions applied in photodetectors are formed between two kinds of semiconductors, or a semiconductor and a conductor (metals or conductive carbon materials). For UV photodetectors, normally one of the two materials is responsible for photon absorption and the other helps to form the junction. The creation of a heterojunction where charges are separated, suppresses photocarriers recombination at the interfaces to optimize photodetection with higher responsivity, quantum efficiency, and photo-gain. Presently, the researches on perovskites, graphene, and other materials are emerging in an endless stream and these materials with excellent properties can be used for heterojunction UV PDs. Perovskites are a good choice for optoelectronic devices because of their tuneable bandgaps, large extinction coefficient, and so on. It has been confirmed that the excellent match in the conduction band of perovskite-TiO_2 junctions can effectively promote charge separation. The heterostructure between $MAPbI_3$ quantum dots (QDs) and TiO_2 shows a relative fast and stable response to UV light, and the combining also enhances the long-term stability of perovskite $MAPbI_3$. Another example is the heterojunctions between graphene and wideband gap semiconductors.

Heterostructure type	Active materials	Wavelength [nm]	Bias [V]	Responsivity [mA W^{-1}]	D^* [Jones]	Response speed	
						Rise time	Fall time
Lattice matching	Se/n-Si	350–600	−2	–	–	–	–
	GaN/ZnO	358	0	0.68	–	–	–
	ZnO/Ga_2O_3	251	0	9.7	6.29×10^{12}	100 µs	900 µs
	ZnO/Ga_2O_3	268	−3	–	–	–	–
van der Waals	$NiPS_3$	254	10	126	1.22×10^{12}	3.2 ms	=17.5 ms
	CuBr	345	10	3.17×10^3	1.4×10^{11}	32 ms	48 ms
	ZnO QDs/graphene	340	10	9.9×10^{11}	$>1 \times 10^{14}$	5 s	85.1 s
	$V\text{-}HfS_2$	405	1	–	–	24 ms	24 ms
	FeS_2-PbS/graphene	340	0.1	1.78×10^9	6.46×10^{11}	–	–
	FeS_2/graphene	340	0.1	1.78×10^9	8.76×10^{11}	0.6 s	0.6 s
	Graphene/GaAs	325	0	186	2.63×10^{13}	–	–
Others	PEDOT:PSS/Ga_2O_3/p-Si	240	0	0.029×10^3	–	60 ms	88 ms
	Graphene-β-Ga_2O_3	254	20	39.3×10^3	5.92×10^{13}	94.83 s (4 V)	219.19 s (4 V)
	$MAPbI_3/TiO_2$	350	1	1.3×10^3	2.5×10^{12}	2 s	1 s
	Si/ZnO	325	−0.04	13	–	19 µs	22 µs
	PEDOT:PSS/ZnO	325	0	3.5	7.5×10^9	5.8 ms	7.3 ms
	$BiOCl/TiO_2$	350	−5	41.94×10^3	1.41×10^{14}	12.9 s	0.81 s
	GD:ZnO/ZnO	365	10	1260×10^3	–	6.1 s	2.1 s
	$CuZnS/TiO_2$	350	3	6.4×10^5	–	–	–
	ZnO/PVK	325	0.2	30	–	1.5 s	6 s
	$TiO_2/MAPbI_3$	350	1	1300	2.5×10^{12}	2 s	1 s
	ZnO/Zn_2SnO_4	300	1	–	9.0×10^{17}	47 ms	47 ms

Figure 3
Different kinds of heterojunction UV PDs

Combining wide band gap semiconductors and graphene, which possesses high carrier mobility and high transparency, is very useful for deep ultraviolet detection. The rectifying effect between graphene and semiconductors such as GaN and Ga_2O_3 leads to promoted responsivity and sensitivity of solar blind ultraviolet detectors. These materials have brought diverse choices of light absorption part of a heterojunction/heterostructure, and provided more methods for enhanced photodetection performance.

Heterojunctions used to be produced by combining semiconductors with different bandgap energies but closely matched lattice parameters. Above the influence of interface defects and surface states, lattice-matched heterojunctions can achieve high separation efficiency and easy transport of photogenerated electron–hole pairs. For example, type II Se/Si heterojunctions with matched lattice own a very small dark current for high sensitivity. Such assembling needs strong chemical bonds that are not easy to achieve, while in van der Waals (vdWs) heterostructures, different materials are assembled physically through weaker vdWs force. VdWs integration frees heterojunction growth from strict requirements of similar lattice characteristics and electrical properties. Therefore, it provides a flexible method that is possible to combine materials with different structures and properties.

In a heterojunction UV PD, if one material is set as UV active materials, the other materials may also have response to visible or even infrared light. Under UV irradiation, photoinduced charge carriers can be generated in both materials. With the help of inner electric fields, these charge carriers can be efficiently separated and transported to electrodes, leading to enhanced UV photo-response. If the heterojunctions are formed by two narrow bandgap materials with response to visible or even infrared light, the as-fabricated heterojunction PDs can exhibit broadband photo-response, in which UV response can be enhanced because of the formed electric field at interfaces. To realize UV selective detection, UV-pass filters can be equipped to eliminate the influence of visible photo-response.

Since heterojunction UV photodetectors are receiving increasing attention for its excellent properties, some future applications are also investigated. $MAPbI_3$ QDs decorated TiO_2 NTs are promising for flexible PDs when constructed on mica substrates through simple spin coating methods. On rigid substrates the heterojunction represents excellent photodetection ability, and when substrates are changed for mica, the performance shows good stability. The device shows high flexibility with stable optoelectronics performance, as well as high transparency. The photocurrent stays almost unchanged when bending at 0°, 45°, 90°, 180° and keeps stable after 200 cycles when bending to 90°. A solar-blind photodetector is constructed on a graphene– diamond heterojunction. The microcrystalline diamond can be easily peeled off from Si substrates because of internal residual stress resulting from lattice mismatching. Thus, applying the diamond thin film to tapes would complete a flexible PD. Monolayer graphene is transferred to the surface of diamond/Au electrode/tape film through wet-transfer method. This flexible junction shows high responsivity under stimulation of 220 nm illumination because of the photo-gain resulting from the defects at graphene/diamond interface

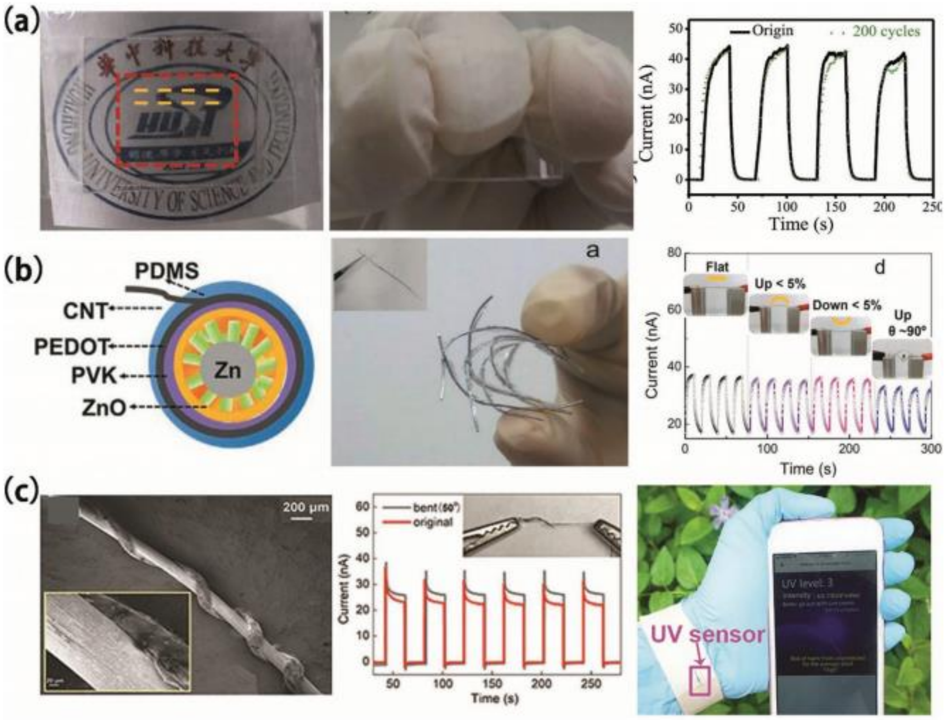


Figure 4
a) Optical images and I–t curves before and after bending of the flexible and transparent heterojunction based on $MAPbI_3$ QDs decorated TiO_2 NTs. b) Schematic representation, optical images, and I–t curves at different bending states of ZnO/PVK PD. c) SEM image, on-off switching tests at different states and applications of CuZnS/TiO_2 PD.

(J. Chen et al., 2020)

Commercially available UV detectors

Si/SiC based detectors:

The most common commercial technical solutions for UV photodetection are mainly Si- or SiC-based, followed on the market by compound semiconductors such as GaAs, GaP and GaAsP. Mainly Schottky barrier photodiodes are fabricated on these materials. This special range of devices notably fits very well for VUV detection applications, for which they offer a remarkable operating stability. SiC photodiodes are one of the best candidates for high-temperature applications. The high temperature operation of a UV sensor had been reported by several groups. The highest operation temperature up to now is 700 °C. The photocurrent of the sensor increased with temperature, and at 400 °C and 700 °C, the photocurrent was approximately double and triple that at RT, respectively. The temperature dependence of the photocurrent reveals the characteristics of absorption particular to indirect transition, and also the minority carrier diffusion length.

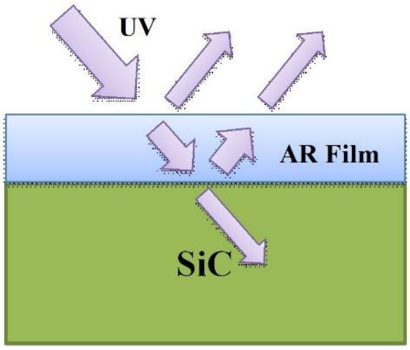


Figure 5
Incidence of UV light from a single-layer AR film to a SiC substrate (multiple reflections in AR films are ignored).

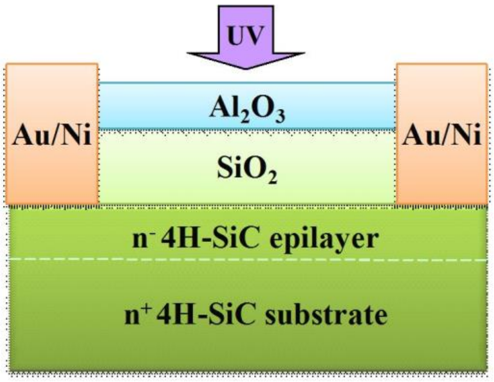


Figure 6
A cross-sectional view of 4H-SiC photodetectors with Al_2O_3 and SiO_2 films deposited by using electron beam evaporation.

(Zhang, 2015)

Nitride (N^{3-}) detectors:

This is one of the most interesting and flexible technical solutions for UV photodetection when considered in the midst of all other semiconductor families available for such application. The first photocurrent characterization studies of polycrystalline Gallium Nitride (GaN) are more than 30 years old. GaN and related compounds are used in a number of applications such as short wavelength LEDs emitting from the green to the blue light, and even reach the UV spectrum, blue lasers, field effect transistors and UV photodetectors. Silicon carbon nitride (SiCN), a wide-bandgap semiconductor, has many interesting physical characteristics such as hardness, high thermal stability, oxidation resistance, and corrosion resistance. Photodetectors based on SiCN film have demonstrated good thermal stability working in the visible-blind region.

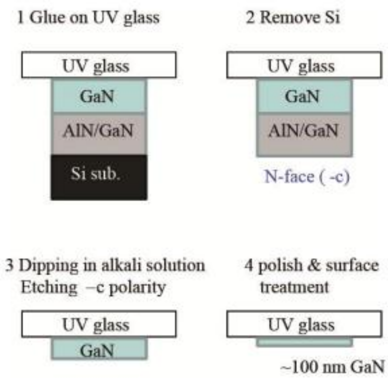


Figure 7
Fabrication process of transparent photocathode based on GaN film grown on

(J. Chen et al., 2020; Omnès et al., 2007; Pandit et al., 2020)

Diamond detectors:

The more recent emergence of diamond-based UV photodetector technologies enables the realization of short cut-off wavelength photoconductors (λ_C=225 nm) associated with a high UV/visible contrast which covers 6 orders of magnitude 1-2. Such devices are extremely suitable for VUV detection applications, for which they offer a remarkable operating stability. The development of this special range of photodetectors remains somehow hindered by the technical difficulty to synthesize high quality monocrystalline films. The final material is therefore in most of the cases a polycrystal whose adjacent grains are significantly misoriented. This results in a large density of electrically and optically active defects which notably degrades the operation of the photoconductors in terms of responsivity and UV/visible contrast. In spite of these problems, diamond-based UV photodetectors are now commercially available, which is the clear indicator of a growing interest for these devices for UV photodetection and its applications.

Figure 8
Diamond detector

(Mainwood, 2000)

Metal Oxide detectors:

Several groups have attempted to enhance the performance of UV photodetectors by developing heterojunction devices consisting of two different metal oxide semiconductors. For example, it was reported the studies of ZnO-Ga_2O_3 core-shell heterostructure UV photodetectors, which demonstrated ultra-high responsivity and detectivity due to an avalanche multiplier effect. A different metal oxide heterojunction, such as NiO/β-Ga_2O_3, was also investigated to provide a high-performance UV photodetector. Firstly, the lattice mismatch of β-Ga_2O_3 and NiO is relatively small. Also, the bandgap of NiO is larger than that of ZnO used in previous study. The p-type behaviour of NiO and n-type β-Ga_2O_3 has led to several reports on the studies of the electrical properties of NiO/β-Ga_2O_3 heterojunction for power electronics applications. Among wide bandgap materials, Ga_2O_3 has many unique characteristics, including high breakdown voltage. These outstanding properties make Ga_2O_3 a promising material for high-temperature and high-power applications. In addition, its bandgap is 4.7–4.9 eV, which is intrinsically suitable for deep ultraviolet (DUV) photodetection without any doping or alloying process. Besides, the growth cost of Ga_2O_3 is relatively low compared to other wide bandgap materials. The basic properties of Ga2O3 and other wide bandgap materials are listed below:

Materials	Si	MgZnO	4H–SiC	GaN	Diamond	β-Ga_2O_3
E_g (eV)	1.1	3.7–7.8	3.3	3.4	5.5	4.8
Thermal Conductivity ($W \cdot cm^{-1} \cdot K^{-1}$)	1.5	1.2	2.7	2.1	10	0.11
Conductive type response	n/p	n/p	n/p	n/p	p	n
Spectrum (nm)	400–1100	200–370	270–380	200–300	<225	<280

(H. Chen et al., 2015; Jia et al., 2020; Omnès et al., 2007; Qin et al., 2019)

Graphene-coated nitride detectors:

Graphene, a two-dimensional (2D) hexagonal lattice array of carbon atoms, is attractive for applications as transparent conductive electrodes because of its high electrical and thermal conductivities, transparency, and mechanical strength. Previous studies have confirmed that the graphene/GaN contact is stable under high temperature and high-radiation operations, indicating that graphene electrodes are appropriate for applications in fire alarms and space exploration devices, which operate in extremely harsh conditions. More previous studies demonstrated the successful implementation of graphene and reduced graphene oxide as transparent electrodes on AlGaN layers with various Al mole fractions, showing sharp cut-of wavelengths at the energy bandgaps of AlGaN. In this regard, the combination of graphene electrodes and AlGaN/GaN heterostructures may yield an excellent UV photodetector, considering the high transparency of graphene in the UV spectral region and the high electrical conductivities of graphene, which would benefit efficient charge collection in the photodetector.

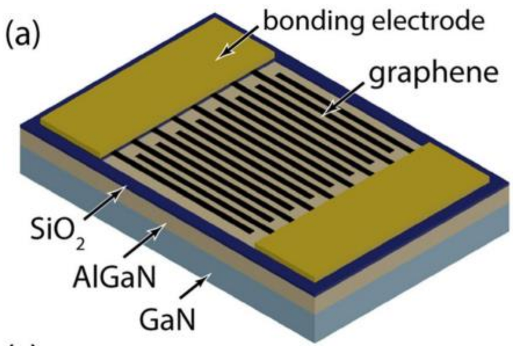


Figure 9
Schematic of the Gr/AlGaN/GaN photodetector structure.

(Luo et al., 2018; Pandit et al., 2020)

One-Dimensional Nanostructured UV Photodetectors:

In the last few years, one-dimensional (1D) or quasi-one-dimensional nanostructures have been widely studied and developed as potential candidates for high-performance UV photodetectors. Their small size allows the nanostructures to exhibit novel and significantly modified physical, chemical, and biological properties, which are different from those of materials in the micro-meter scale. As a result of the large surface-to-volume ratio and small dimensions, the nanostructured materials are especially sensitive to the light (photoconductivity). Moreover, the possibility to integrate functionality in nanostructures, such as homo- and hetero-junctions, within single nanowires or nanobelts, enables large scale integration. Nanostructured ZnO materials have received broad attention due to their distinguished performance in the photonic and electronic field. ZnO has a wide bandgap of 3.34 eV at room temperature (RT), which makes it very suitable for photodetection working in the UV-A region from 320 to 400 nm. An important characteristic of ZnO is its large exciton binding energy (60 meV), which can ensure efficient excitonic emission at RT. The UV luminescence has been reported in disordered nanoparticles and thin films. Due to its wide bandgaps, ZnO is transparent to visible light, and can be made highly n-type conductive by doping. In addition, ZnO nanostructures have unique advantages including high specific surface area, low toxicity, chemical stability, electrochemical activity, and high conductivity. Therefore, they are promising material for sensors applications with a high performance. (Sang et al., 2013)

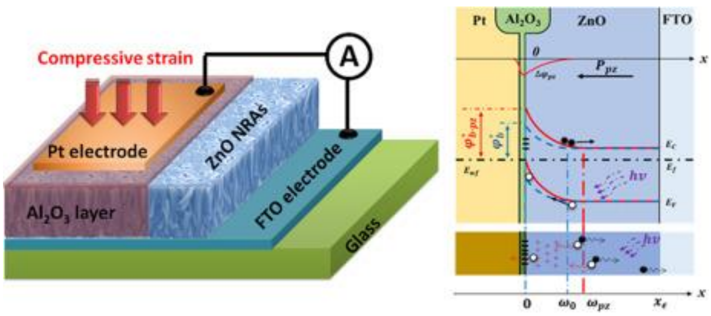


Figure 10
Structure and geometry of the nanorods-based photodetector. (a) A schematic diagram of the photodetector. (b) The energy band diagram of the photodetector at zero bias

Hybrid Photodetectors:

In recent years, to achieve multiple or wide band absorption, or to realize the better spectrum selectivity independently, hybrid photodetectors are attracting more and more attention. Multicolour optical sensing with high sensitivity at designed wavelengths can be applied in a variety of applications, such as imaging, surveillance, optical communication, remote control, and target identification. Since the absorption of organic materials can be easily tuned by tailoring the chemical structure, UV sensors based on those materials seems to possess more flexibility in realizing spectra selective responses. It was demonstrated a low-cost UV sensor based on polymer/ZnO nanorods, and the device showed a narrow-band response in the region of 300–360 nm. The hybrid UV detector composed of TiO_2 nanorods and polyfluorene showed obvious UV photoconductive effect, and the response was fast to the switching on and off of UV light illumination, which was repeated at least 50 times.

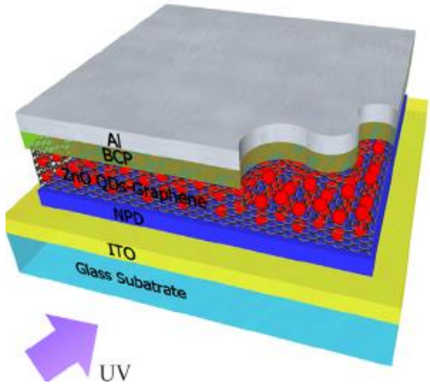


Figure 11
Schematic illustration of the hybrid UV photodetector fabricated from ZnO QDs-graphene composite material and polymers.

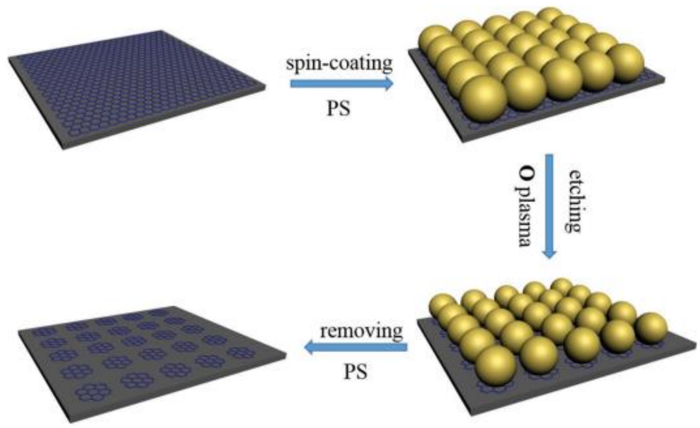
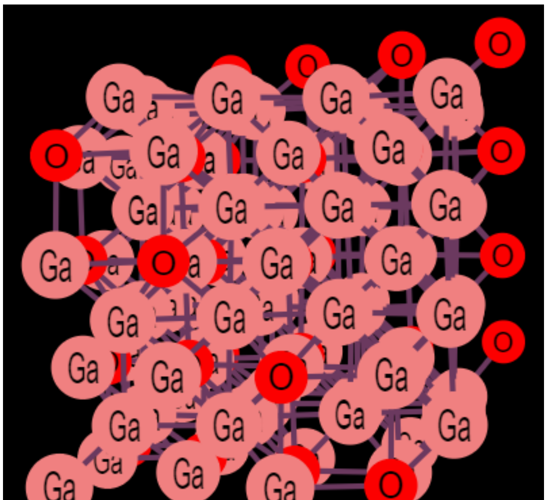


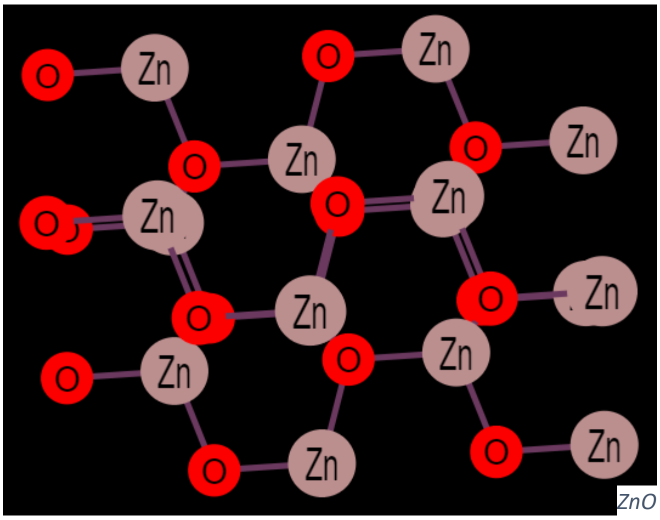
Figure 12 Schematic illustration of the fabrication procedures of the GNDA (graphene nanodot array) based on PS-NS (polystyrene-nanospheres) lithography. First, a closely packed PS NSs layer was assembled on the graphene/silicon wafer substrates by spin-coating. Then RIE (reactive ion etching) was employed to fabricate graphene nanodots array with PS NS layer as mask. During RIE, PS NSs and graphene unprotected by PS NSs were etched by oxygen plasma. As a result, only graphene underneath the remaining PS NSs was left. Finally, the remaining PS NSs were removed selectively, and the orderly aligned GNDA were obtained.

(H. Chen et al., 2015; Donati, 2000; Sang et al., 2013; Shin & Choi, 2018)

Crystal Structures for the above detectors' material



Ga_2O_3



ZnO

Applications

General Applications:

Interest in a new generation of wide band-gap semiconductor detectors stems in part from the fact that the Earth's atmosphere is opaque at wavelength below 300 nm. To take advantage of this optical window, Earth to space communication will need detectors that are UV sensitive but blind to the ambient visible radiation. Another area where these detectors can find applications is in the combustion monitoring of gases where UV emission is a normal by-product. In radiometry and photometry, they can be used for measuring properties like optical power, luminous flux, optical intensity and irradiance, in conjunction with additional means also for properties like the radiance. They are used to measure optical powers, for example in spectrometers, light barriers, optical data storage devices, autocorrelators, beam profilers, fluorescence microscopes, interferometers and various types of optical sensors. Particularly sensitive photodetectors are required for laser rangefinders, LIDAR, quantum optics experiments and night vision devices. Particularly fast photodetectors are used for optical fibre communications, optical frequency metrology and for the characterization of pulsed lasers or laser noise. Mostly two-dimensional arrays containing many identical photodetectors are used as focal plane arrays, mostly for imaging applications. For example, most cameras contain such devices as image sensors.

(Donati, 2000; L.C. Passos & M.F.S. Saraiva, 2019; *Photodetector - Wikipedia*, n.d.; Razeghi & Rogalski, 1996)

UV-Visible Absorption spectroscopy:

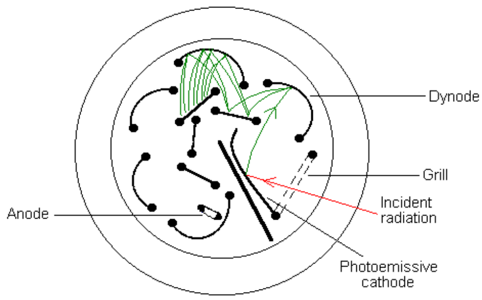


Figure 13
Cross section of a photomultiplier tube

Photomultiplier tube is a commonly used detector in UV-Visible spectroscopy. It consists of a photo-emissive cathode (a cathode which emits electrons when struck by photons of radiation), several dynodes (which emit several electrons for each electron striking them) and an anode.
A photon of radiation entering the tube strikes the cathode, causing the emission of several electrons. These electrons are accelerated towards the first dynode (which is 90V more positive than the cathode). The electrons strike the first dynode, causing the emission of several electrons for each incident electron. These electrons are then accelerated towards the second dynode, to produce more electrons which are accelerated towards dynode three and so on. Eventually, the electrons are collected at the anode. By this time, each original photon has produced 106 - 107 electrons. The resulting current is amplified and measured. Photomultipliers are very sensitive to UV and visible radiation. They have fast response times. Intense light damages photomultipliers and are therefore limited to measuring low power radiation.
Linear photodiode array is an example of a multichannel photon detector. These detectors are capable of measuring all elements of a beam of dispersed radiation simultaneously. A linear photodiode array comprises many small silicon photodiodes formed on a single silicon chip. There can be between 64 to 4096 sensor elements on a chip, the most common being 1024 photodiodes. For each diode, there is also a storage capacitor and a switch. The individual diode-capacitor circuits can be sequentially scanned.
Charge-Coupled Devices (CCDs) are similar to diode array detectors, but instead of diodes, they consist of an array of photo-capacitors.
(L.C. Passos & M.F.S. Saraiva, 2019; *UV-Vis Absorption Spectroscopy - Instrumentation*, n.d.)

Smoke and fire Detectors:

Flame detectors alarm at the presence of light from flames, usually in the ultraviolet or infrared range. The detectors are set to detect the light flicker of a flame. They may be equipped with time-delay features to eliminate false alarms from transient flickering light sources. There are six types of optical detectors commonly used:

1. Ultraviolet (UV)
2. Single-frequency infrared (IR)
3. Dual-frequency infrared (IR/IR)
4. Ultraviolet/infrared—simple voting (UV/IR)
5. Ultraviolet/infrared—ratio measurement (UV/IR)
6. Multiband.

Figure 14
Flame Detector

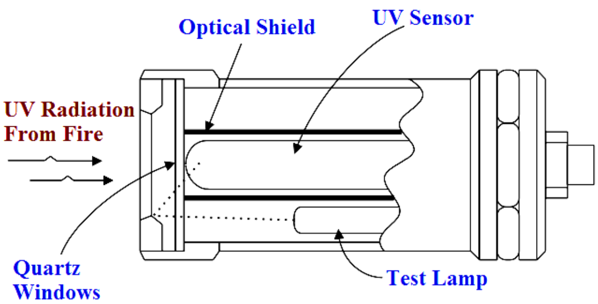


Figure 15
Schematic diagram of a UV flame detector

UV detectors (which is a subject of this report) respond to the relatively low energy levels produced at wavelengths between 185nm and 245nm. This wavelength is outside the range of normal human visibility and outside that of sunlight. It responds to most burning materials but at different rates. The detector can be extremely fast, i.e., less than 12ms for special applications (e.g., explosive handling). It is generally indifferent to the physical characteristics of flames and does not require a “flicker” to meet signal input functions. It is not greatly affected by deposits of ice on the lens but can be affected by deposits of grease and oil on the lens. This reduces its ability to sense a fire. Some vapours, typically those with unsaturated bonds, may cause signal attenuation. Smoke will cause a reduction in signal level seen during a fire. Even with all these limitations, the UV detector is a general all-purpose detector and special modules are available that can be used in high-temperature applications up to 125˚C.

(Nolan, 2019)

Chromatography detectors:

1) Gas Chromatography:

Analytical performance characteristics of a new vacuum ultraviolet (VUV) detector for gas chromatography (GC) are reported along with other detectors like FID (Flame Ionisation Detector) and TCD (Thermal Conductivity Detector) but are currently under study. GC-VUV can be applied to hydrocarbons, fixed gases, polyaromatic hydrocarbons, fatty acids, pesticides, drugs, and oestrogens. Applications of these new detectors are chosen to feature the sensitivity and universal detection capabilities of the VUV detector, especially for cases where mass spectrometry performance has been limited. Quantitative analysis is governed by Beer−Lambert Law relationships.
Prior efforts aimed at the development and integration of UV and VUV absorption detection with GC have been limited. A far UV detector (FUVD) was first combined with capillary GC in

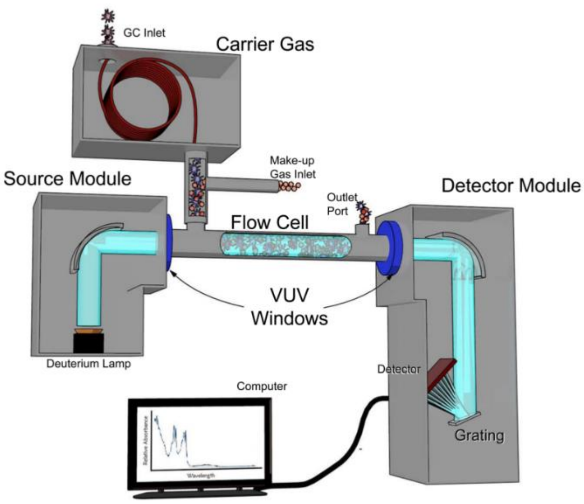


Figure 16
Schematic of the GC-VUV instrument

1987. It was limited to probing essentially a single wavelength absorption at 122 nm (10.2 eV), which is sufficient for detection of a wide variety of chemical species including alkanes, but provided no qualitative information. A more energetic photoionization detector (PID) (106 nm; 11.7 eV) was also evaluated in that study. Originally commercialized in 1976, the higher energy PID showed less interference from compounds, such as water compared to the 10.2 eV FUVD.

(Schug et al., 2014)

2) High Performance Liquid Chromatography (HPLC):

In HPLC, UV-visible detectors are the most commonly used detectors for trace analysis of UV-absorbing solutes. Low-wavelength UV detection gives better sensitivity than other sensitive detectors (for example, refractive index detector). It is relatively insensitive to temperature changes and mobile phase flow, easier to operate and is non-destructive to the sample. For these reasons and many more, UV detectors are very reliable in HPLC over other detectors. There are various settings in which the detectors can be made according to the use, convenience and overall to increase versatility of the detectors giving rise to different types of UV detectors.

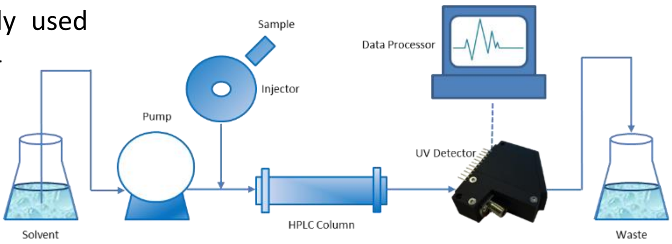


Figure 17
Common HPLC setup

Types of UV detectors in HPLC:

1. **Fixed wavelength detector** where a light of a fixed wavelength of 254nm is shone on the reference and sample cells and the UV absorption is detected. These detectors were the mainstay of UV detection prior to the introduction of the variable and diode array detectors, but they are not widely used today. Their current appeal is low price and simple construction, and they tend to be more popular in the educational environment or other budget-limited settings.

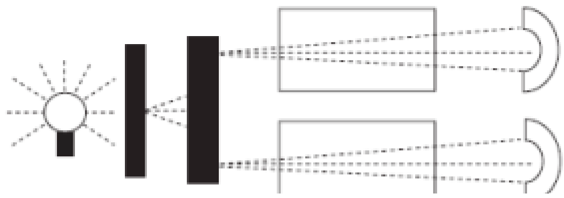


Figure 18
Fixed wavelength detector

2. **Variable wavelength detector** where a broad-spectrum UV lamp (typically deuterium) is directed through a slit and onto a diffraction grating. The grating spreads the light out into its component wavelengths, and the grating is then rotated to direct a single wavelength (or narrow range of wavelengths) of light through the slit and detector cell and onto a photodetector. These detectors usually use a sample and reference cell configuration for differential detection.

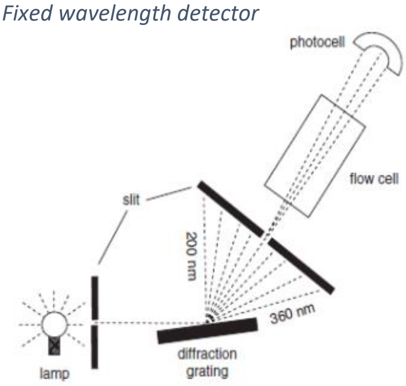


Figure 19
Variable wavelength detector

3. **Diode array detector** which has a similar optical path to the variable-wavelength detector, except that the white light from the lamp passes through the flow cell prior to striking the diffraction grating. This allows the grating to spread the spectrum across an array of photodiodes, hence the name diode array. The number of photodiodes varies with the specific brand and model of detector, but detectors with 512 or 1024 diodes are common.

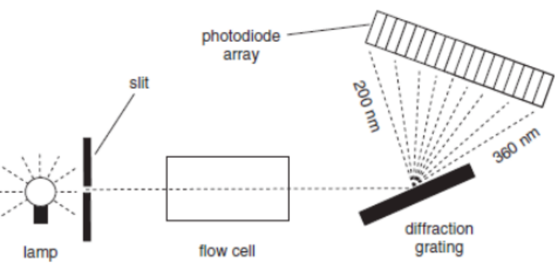


Figure 20
Diode array detector

Case Study:

Graphene, Existing and Upcoming Technologies

Introduction: Graphene

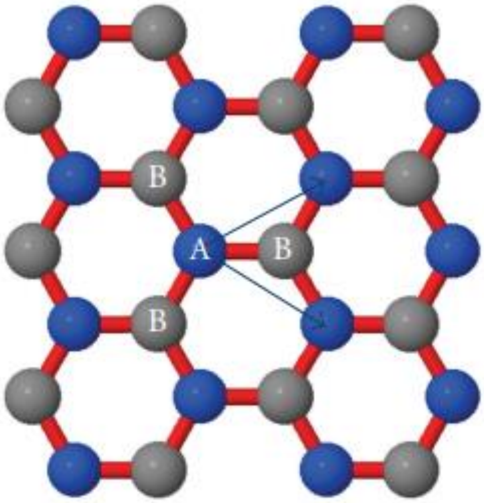


Figure 21
Triangular sublattices of graphene. Each atom in one sublattice (A) has 3 nearest neighbours in sublattice (B) and vice versa.

Graphene is the name given to a two-dimensional sheet of sp^2 -hybridized carbon. Its extended honeycomb network is the basic building block of other important allotropes; it can be stacked to form 3D graphite, rolled to form 1D nanotubes, and wrapped to form 0D fullerenes. Long-range π-conjugation in graphene yields extraordinary thermal, mechanical, and electrical properties, which have long been the interest of many theoretical studies and more recently became an exciting area for experimentalists. Atomic planes are constituents of bulk crystals, but one-atom-thick materials such as graphene remained unknown. The basic reason for this is that nature strictly forbids the growth of low dimensional crystals. Crystal growth implies high temperatures and, therefore, thermal fluctuations that are detrimental for the stability of macroscopic 1D and 2D objects. Graphene is a wonder material with many superlatives to its name. It is the thinnest known material in the universe and the strongest ever measured. Its charge carriers exhibit giant intrinsic mobility, have zero effective mass, and can travel for micro-meters without scattering at room temperature. Graphene can sustain current densities six orders of magnitude higher than that of copper, shows record thermal conductivity and stiffness, is impermeable to gases, and reconciles such conflicting qualities as brittleness and ductility. Electron transport in graphene is described by a Dirac-like equation, which allows the investigation of relativistic quantum phenomena in a benchtop experiment. Graphene research has developed at a truly relentless pace. Several papers appear every day, and, if the bibliometrics predictions are to be trusted, the amount of literature on graphene will keep rapidly increasing over the next few years.

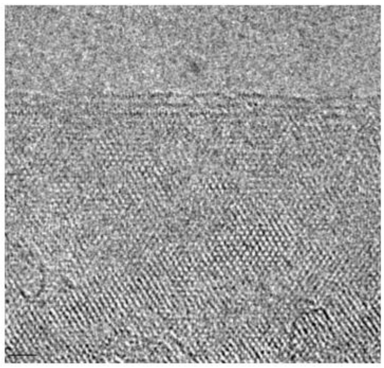


Figure 22
Atomic scale TEM image of suspended graphene.

Graphene has a remarkable band structure thanks to its crystal structure. Carbon atoms form a hexagonal lattice on a two-dimensional plane. Each carbon atom is about a = 1.42A from its three neighbours, with each of which it shares one σ-bond. The fourth bond is a π-bond, which is oriented in the z-direction (out of the plane). One can visualize the π orbital as a pair of symmetric lobes oriented along the z-axis and centred on the nucleus. Each atom has one of these π-bonds, which are then hybridized together to form what are referred to as the π-band and π*-bands. These bands are responsible for most of the peculiar electronic properties of graphene. The hexagonal lattice of graphene can be regarded as two interleaving triangular lattices. This is illustrated in Figure 16. **Graphene versus Traditional Materials:** Below summarized is some of the interesting properties of graphene by comparing them with more traditional materials such as 2D semiconductors:

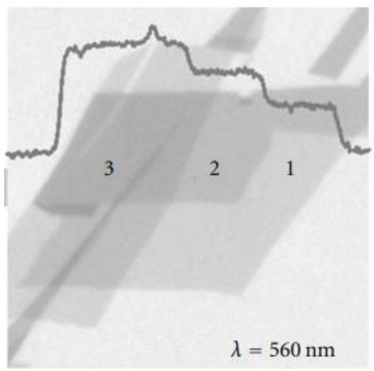


Figure 23
Optical microscope image at 560nm of graphene.

1. Traditional semiconductors have a finite bandgap while graphene has a nominal gap of zero.

2. Dispersion in graphene is chiral. This is related to some very distinctive material behaviours like Klein tunnelling.
3. Graphene has a linear dispersion relation while semiconductors tend to have quadratic dispersion. Many of the impressive physical and electronic properties of graphene can be considered to be consequences of this fact.

Optical Properties: The properties of graphene make it an attractive choice for use in optoelectronic devices. In particular, graphene's high transparency, low reflectance, high carrier mobility, and near-ballistic transport at room temperature make it a promising choice for transparent electrodes. Optically, single-layer graphene (SLG) has an unusually high absorption (given its thickness) that can be described in terms of the fine structure constant, α (a fundamental physical constant that describes the interaction of matter and electromagnetic fields). Because graphene sheets behave as a 2-dimensional electron gas, they are optically almost non-interacting in superposition (though the same cannot be said of their electronic interaction), so the absorbance of few-layer graphene (FLG) sheets is roughly proportional to the number of layers. The absorption spectrum of graphene from the ultraviolet to infrared is notably constant around 2- 3% absorption, as shown in Figure 19, compared to some other materials.

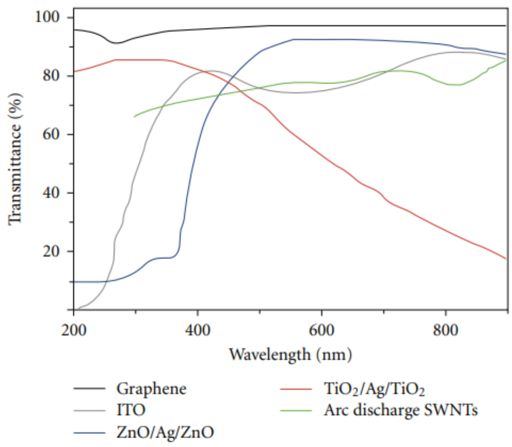


Figure 24
The absorption spectrum of graphene from the ultraviolet to infrared compared to some other materials.

Despite the absence of a bandgap in graphene, it can function as the semiconductor active element for a metal/graphene photodetector. Normally, photoexcitation of graphene results in electron-hole pairs that quickly recombine. In graphene, both electrons and holes have high mobility, providing an advantage over conventional semiconductors. Graphene has found its way into many optoelectronic and photonic devices, a portion of which will be covered here. Other uses, including photodetectors, touch screens, smart windows, saturable absorbers, and optical limiters.

(Allen et al., 2010; J. Chen et al., 2020; Geim, n.d.; Shimomura et al., 2021; Wang et al., 2020; Xia et al., 2009; Yang et al., 2018)

Graphene and Graphene-based Photodetectors

Graphene photodetectors are highly attractive owing to its ultra-fast and wide-range spectral response from visible to infrared benefit from the superior carrier mobility and the linear dispersion with zero bandgap of graphene. It is a gapless semiconductor, allowing light absorption over a very wide wavelength spectrum ranging from ultra-violet to terahertz frequency radiations. Along with that, graphene also exhibits extremely high carrier mobility up to ~106 cm^2 /V·s and is capable for ultra-fast operation. Most graphene photodetectors utilize graphene-metal (or semiconductor) junctions or graphene p-n junctions to spatially separate and extract photon generated carriers. However, the reported maximum responsivity of photodetectors based on the above-mentioned pure graphene junctions is lower than 10 mA/W. Monolayer graphene absorbs 2.3% of the incident light, remarkably high for a material with only one atom thickness, but still very low in absolute terms. Many efforts

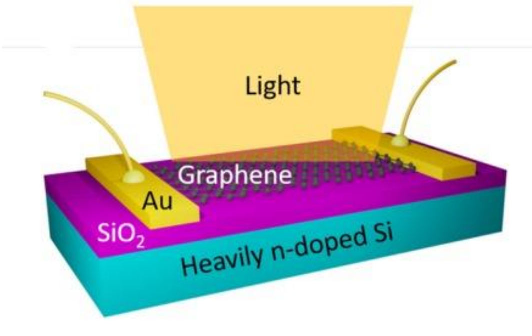


Figure 25
Schematics of a graphene photodetector

have been explored to improve the performance of graphene photodetectors. For example, hybrid graphene-quantum dots photodetectors have shown significantly enhanced responsivity. However, the bandwidth and response time of such hybrid photodetector are limited by the narrow spectral width which is determined by the quantum dots rather than by graphene.

Light can be detected by the generation of a photovoltage at the graphene/electrode interface because of electron–hole separation by graphene band bending in the vicinity of the two electrodes. Since the polarities of the photovoltages for these two electrodes are opposite to each other, the photovoltages at the two graphene/electrode interfaces are cancelled out under simultaneous light irradiation of these electrodes. Hence, a low photo signal can be obtained in typical graphene-based photodetectors for detecting macroscopic light, which has a spot size that is much larger than the graphene size. Significant effort has been put into preventing the photovoltage cancelation in previously developed graphene photodetectors. One method is local light irradiation of one side of the graphene/electrode interface with an objective lens or a silicon waveguide to avoid the cancelation of the photovoltage; however, the problem is that a large optical system or fine device structure based on silicon photonics are required for the local light irradiation.

Since, single-layer graphene has low light absorbance (only 2.3%) in the ultraviolet (UV) to near infrared (NIR) region and short light-matter interaction length, unfavourable for light harvesting applications and the ultra-short lifetime of excitons in pure graphene resulting from its gapless nature also leads to fast carrier recombination, which limits the efficient production of photovoltage, the emergence of the graphene/SiC interface in 2009 as the prototype of graphene/semiconductor heterostructure has attracted much attention due to the expected synergistic properties of the two materials. Afterwards, the rapid development of graphene transfer techniques has led to a wide variety of unique designs for the functionality of the device geometry based on graphene/semiconductor hybrid heterostructures. Unlike other detector structures, the best feature of graphene/semiconductor junctions is the adjustable Schottky barrier height, useful not only for understanding the interface transport mechanism, but also for adjusting the device functionalities. Recent studies have shown that graphene/semiconductor interfaces/heterojunctions are efficient for generation, separation, and transmission of photocarriers, thereby exhibiting new features in their optoelectronic applications.

(Allen et al., 2010; Cakmakyapan et al., 2018; Cooper et al., 2012; Guo et al., 2016; Murali et al., 2019; Shimomura et al., 2021; Xia et al., 2009)

Graphene dopants

Graphene/Si: The built-in electric potential at the graphene/Si interface is determined by the difference between the work functions of graphene and Si when no bias is applied. Pristine graphene forms a low Schottky barrier with Si due to the relatively-small work function (4.5 eV), and the pristine graphene/Si junction exhibits a large series resistance due to the relatively large sheet resistance of pristine graphene. These issues should be overcome to enhance the performance of the graphene/Si PDs. Chemical doping is a very efficient approach for increasing the work function and conductivity of graphene, resulting in the

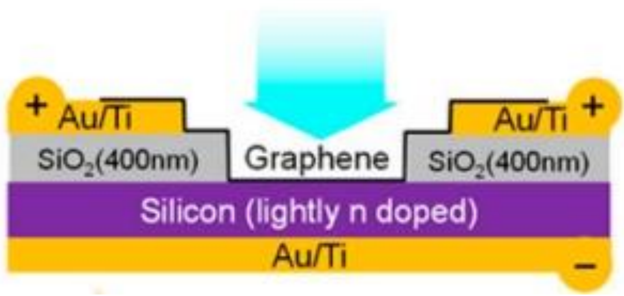


Figure 26
Schematic diagram of a typical monolayer graphene/Si heterojunction.

increase of the Schottky barrier and the reduction of the series resistance. The electrons and holes are then less recombined, thereby improving the performance of the PDs.

(Luo et al., 2018; Shimomura et al., 2021)

<u>Graphene/GaN:</u> GaN is a wide direct bandgap semiconductor with excellent optical/electrical properties and high chemical stabilities at high temperature, and has been employed in various PD applications. Graphene/GaN heterostructures are useful for light detection in the UV/visible region based on Schottky diode junction. Graphene-TCE based GaN Schottky diode for UV detectors has shown excellent performance of high I_{light}/I_{dark} ratio under UV illumination. In another approach, UV PDs were successfully fabricated by transferring large-area graphene TCE (transparent conductive electrodes) on vertically-standing GaN NW array as a light absorbing media, as shown in Figure

(Guo et al., 2016; Xia et al., 2009)

<u>Graphene/ZnO:</u> ZnO has received considerable attention over the past two decades due to its unique properties such as direct bandgap of 3.2 eV, large exciton binding energy of ~60 meV, high transmittance (>80%) in the wavelength range of 400–800 nm, and wide-variable conductivity in the metallic to insulating range. To date, graphene/ZnO heterostructures have been extensively studied for PD application. In another approach, novel ZnO-QDs-based heterostructures were combined with graphene layers for PDs to show excellent visible-blind UV photo-response, resulting from the reduction of the DC, caused by the formation of the potential barriers between adjacent ZnO QDs and between graphene and ZnO QDs. Under UV irradiation, the holes separated from the electrons under photoexcitation are captured by the surface states to discharge the absorbed oxygen ions on the surface of the ZnO QDs. The unpaired electrons with a relatively long lifetime move to the graphene layer and then transport in the graphene channel, leading to modulation of the transport characteristics of graphene. When UV light is turned off, the surrounding oxygen molecules are re-adsorbed on the QD surface, thereby trapping electrons and lowering the Fermi energy level, resulting in the recovery of the transport characteristics of the graphene.

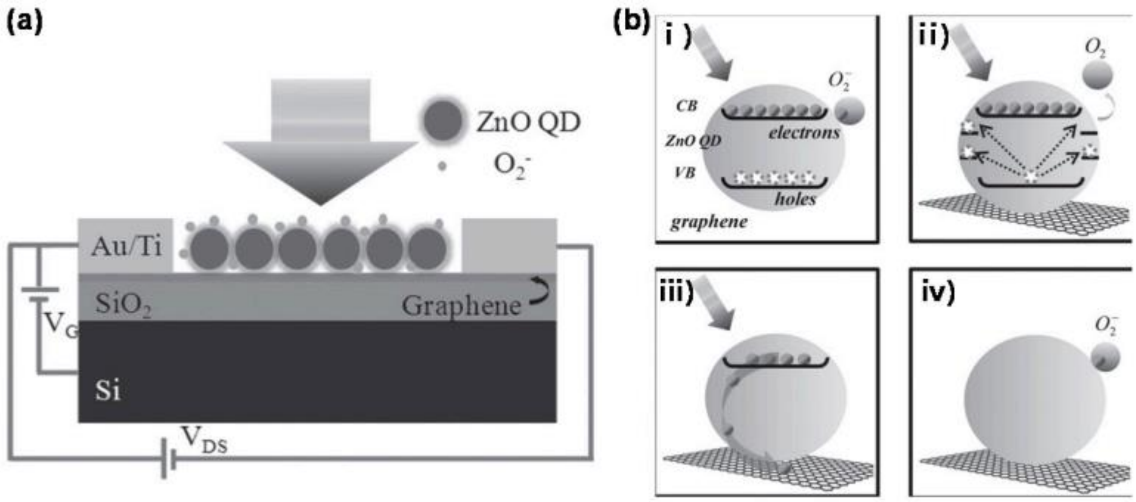


Figure 27
(a) Schematics of the graphene device coated with ZnO QDs; (b) The mechanism of the oxygen-assisted charge transfer process. (i) With irradiation of photon energy larger than the bandgap of the QDs, electron-hole pairs are generated; (ii) The holes are trapped at the surface states, leaving behind the unpaired electrons. The trapped holes will discharge the oxygen ions on the surface, and the oxygen molecules are desorbed; (iii) The electrons will transfer from the QDs to the graphene layer; (iv) With laser turned off, no more electron-hole pairs are generated. The oxygen molecules will re-adsorb on QDs and capture the remaining electrons to form oxygen ions on the surface

(Fu et al., 2012; Tang et al., 2018)

Graphene/Perovskites: Perovskites have recently received strong attention as next-generation PD materials because of their high adsorption coefficient, tuneable bandgap, small exciton binding energy, long carrier diffusion length, ambipolar charge transport, and high carrier mobility. The perovskite material has a specific crystal structure as ABX_3 formula. The larger A cations occupy cuboctahedral sites shared with twelve X anions, while smaller B cations stabilize at octahedral sites shared with six X anions. Organic-inorganic halide perovskite materials are commonly used in solar energy applications. The graphene/perovskite hybrid structure is very useful for taking advantage of the synergy effect based on the unique properties of graphene and perovskite materials. Figure shows a typical early-stage phototransistor composed of graphene/$CH_3NH_3PbI_3$ ($MAPbI_3$) hybrid, exhibiting the R of several hundreds of A/W and relatively-rapid response time shorter than 1s in the UV-visible range. Especially, Figure d shows on-off switching behaviours of the perovskite-graphene PD with rise and fall times of 87 and 540ms, respectively. These results show the robustness and reproducibility of the device.

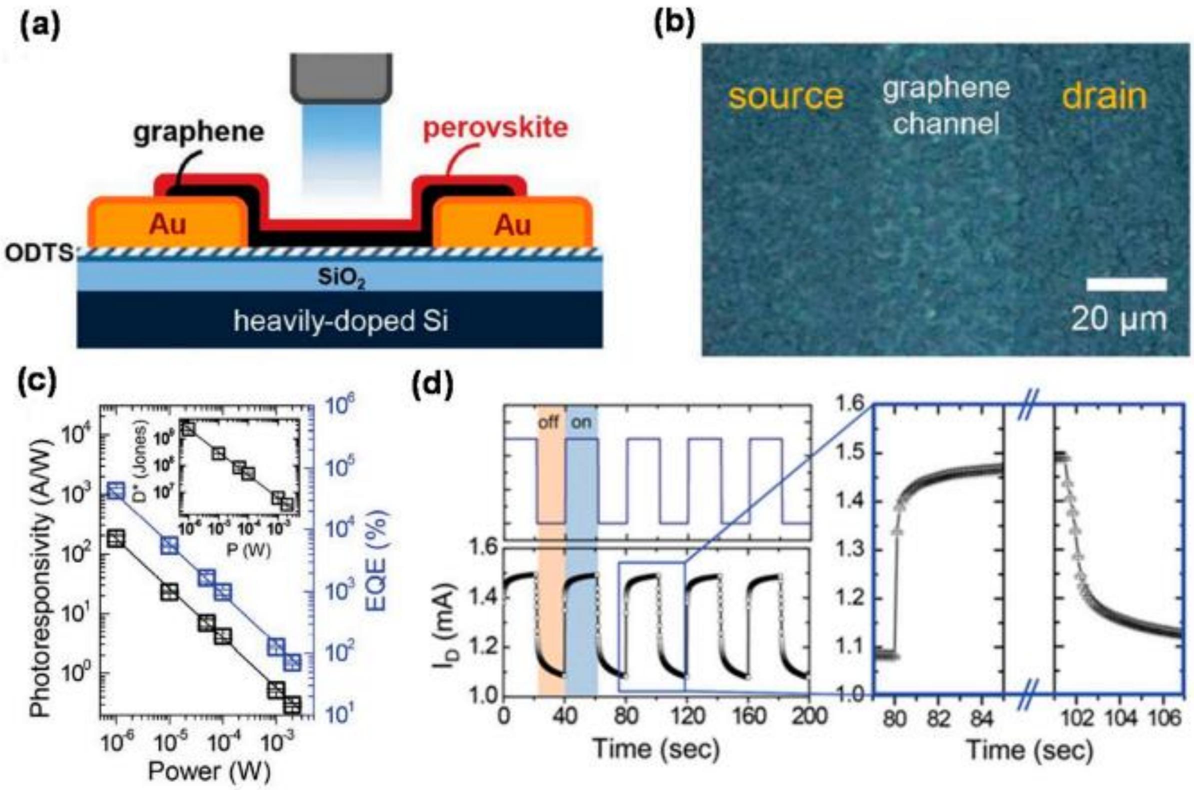


Figure 29
a) Schematic diagram of the perovskite-graphene hybrid PD; (b) Optical microscopy image of the $CH_3NH_3PbI_3$-graphene PD; (c) Photoresponsivity and EQE vs. illumination power. The inset shows the photo-detectivity (D) vs. illumination power; (d) On-off switching characteristics of the PD under dark and light illumination. By the pulsed laser illumination (left top), the temporal response of the photocurrent is shown (left bottom).*

(Shin & Choi, 2018)

Graphene/poly-BrNpA: The hybrid graphene/poly-BrNpA photodetector takes the FET (Field Effect Transistor) configuration. Except for radiative (fluorescence, lifetime of ns; phosphorescence, lifetime of ms) and non-radiative transitions, the electron-hole pairs, photo-induced in the poly-BrNpA film, have a long lifetime due to electron transfer to graphene. The photo-induced holes are trapped in the poly-BrNpA film. As a result, there emerges a built-in photogate at the interface between the poly-BrNpA film and graphene. The electrons transferred into graphene participate in the generation and multiplication of photocurrent under a certain drain-source bias thanks to the photogating effect. In the ambient condition (~45% humidity), the hybrid graphene/phosphor photodetector has a UV response relying on the optical intensity. When the optical intensity of UV irradiation is far beneath that to stimulate photoisomerization, photocurrent forms due to the photogating effect with additional electrons in graphene transferred from the photo-generated electron-hole pairs in the poly-BrNpA film. The photodetector has a gain due to slow recombination of the flowing electrons in the circuit with the photo-generated holes trapped in the phosphor layer.

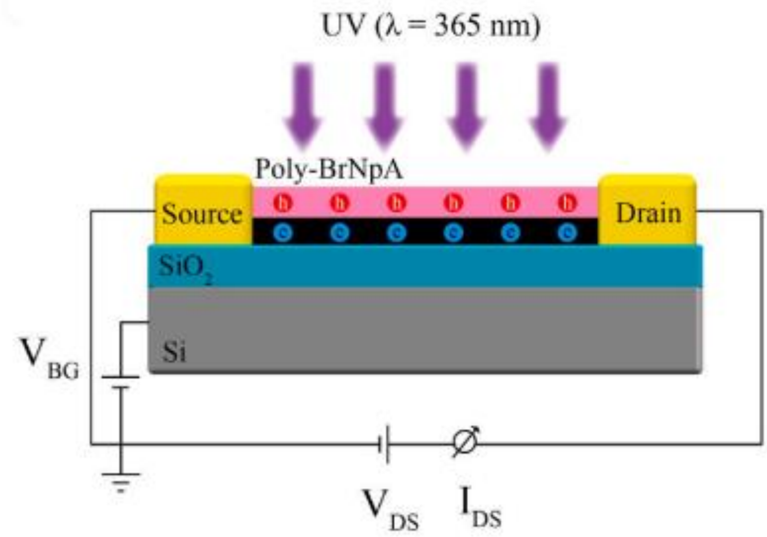


Figure 30
Schematic diagram of the photocurrent generation for the hybrid graphene/poly-BrNpA photodetector under UV irradiation at 365 nm

(Li et al., 2020)

Future of Graphene and Graphene-based photodetectors:

Graphene's journey is quite remarkable considering the tremendous scientific and technological impact this material has had on the scientific community. It has an interesting history, but many now wonder about its future. The subject of considerable scholarly debate, it does seem reasonable to assert a few things looking ahead. First, the quality and availability of "synthetic" graphene will continue to improve. Whether high quality material comes in the form of an alternative chemical route to the complete exfoliation of graphite or from optimization of the thermal processes required for substrate-based methods, there is no sign that synthetic techniques are nearing their upper limit. This means that device engineers will have ample access to improved materials for developing novel structures and finding ways to integrate graphene into present-day electronic devices. Second, chemical modification of graphene's basal plane or its edges will substantially influence graphene-based devices. For electronic applications, one can imagine the attachment of functional groups aimed at self-assembly of simple circuits or the incorporation of chemical dopants to limit leakage current under zero gate bias. For sensors, lockand-key type binding sites could provide selective sensitivity to a wide variety of analytes. These might include chemical warfare agents or even biological species. Third, industrial use of graphene as a transparent conductor could have huge implications for the solar industry. As synthetic routes improve, the prospect of replacing ITO (Indium Tin Oxide) with a low-cost carbon-based coating seems feasible. This would not only remove significant uncertainty about the availability and cost of indium but also enable non-evaporative roll-to roll processing of transparent conductors. Research on graphene's electronic properties is now matured but is unlikely to start fading any time soon, especially because of the virtually unexplored opportunity to control quantum transport by strain engineering and various structural modifications. Even after that, graphene will continue to stand out in the arsenal of condensed matter physics.

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