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*Review Article*

# Recent Advances in Indium Selenide (InSe) based Photodetectors: A Mini Review

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Indium Selenide (InSe) is an emerging two-dimensional (2D) layered metal monochalcogenide (MMC) material which is highly regarded for its unique material properties. Due to the large surface area, high electron mobility and bandgap tunability (Visible to IR), InSe is widely sought for polarization sensitive photodetection. In the recent years, InSe heterostructure based broadband photodetectors (UV-Vis-IR) have received significant scientific attention. Photodetectors based on InSe layers/flakes and their heterostructures (with oxides, graphene, TMDCs, *etc.*) have displayed ultrahigh efficiency, fast switching and self-powered operation. Till now, a record breaking photoresponsivity up to$10^7$ $AW^{-1}$ with switching time less than 2 μs for InSe based photodetector has been reported. Though, despite of scientific advancements, InSe based photodetectors suffer from numerous technological challenges. Therefore, in this mini review, we present a systematic and comprehensive review of noteworthy recent developments, scientific and technological challenges of InSe based optoelectronic devices. A brief discussion on the future aspects of InSe based photodetectors has also been presented.

**Keywords**: UV-Vis-IR; Photodectors; Graphene; Thin Film

## 1 Introduction

Thin film based photodetectors play a great role in modern day telecommunication, imaging (biomedical, optical *etc.*), spectroscopy, remote control and other consumer electronics[1-4]. Since the discovery of graphene in 2004, two-dimensional (2D) materials owing to small architecture, high mobility and broadband photo absorption and have been explored extensively for optoelectronic applications[3]. As the development of graphene based photodetectors was limited due to its zero bandgap, researchers found promising potential in layered transition metal dichalcogenides (TMDs). TMD material systems with a formula $MX_2$ (M: Mo, W, Ga, In and X: S, Se, Te) have emerged as a substitute of conventional 2D semiconductors for broadband photodetection[2-4].

In recent years, Indium Selenide (InSe) has appeared as a successor of layered van der Waals (vdW) based TMD material, with promising potential in the field of nano- and opto-electronics[1]. Due to their exceptional electronic properties, *i.e.*, large carrier mobility ($>10^3$ $cm^2V^{-1}s^{-1}$), small effective mass ($m^* = 0.143m_o$) and layer dependent bandgap tunability (1.2 - 2.3 eV), InSe based photodetectors have shown superior photo-conducting performance[2]. It has been witnessed that photodetectors fabricated using InSe heterojunctions has exhibited broadband photo response (400-1000 nm), ultrafast switching(up to 2 μs) and high detectivity[3]. Recent studies have revealed that InSe thin films with exceptional field effect mobility have displayed extraordinary photo-responsivities ($\sim10^6$ - $10^7$ $AW^{-1}$) which is much higher than other non-graphene contacted 2D semiconductors based photodetector[4].

As this area of research is relatively new and unexplored, researchers have encountered numerous roadblocks in optimizing the performance of InSe based optoelectronic devices as well. Although significant progress in the development of InSe based photodetectors has been made in the past few years, it is just the beginning to realize a matured device technology. Therefore, this mini review emphasizes and reports some of the notable recent advancements in the field of InSe based photodetectors. Further, challenges associated with material properties, device fabrication, efficiency optimization and technology upgradation have also been discussed briefly.

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## 2 Material Synthesis

InSe is a III-VI group based two dimensional (2D) metal monochalcogenide (MMC) material with strong interlayer interaction containing covalently bonded Se-In-In-Se layers. The fundamental properties of InSe *i.e.*, bandgap, mobility etc. has been observed to be altered with an increase/decrease in layer thickness[1-4].

At present, 2D InSe layers are synthesized using various techniques in the form of thin films, flakes, nanostructures *etc*. Among them, mechanical exfoliation of bulk InSe crystalsis the most sought technique to synthesize InSe layers or flakes[5]. The exfoliated InSe flakes, thin films or nanostructures are then transferred on intended substrates and used for the fabrication of standalone or heterojunction based photodetectors[5,6]. While standalone InSe based photodetectors absorb a certain region of the electromagnetic spectrum, InSe heterojunction based photodetectors with tailored properties offer optimized performance such as broadband photodetection and self-powered operation[4]. Though, despite such high performance, the mechanical exfoliation technique also suffers from numerous challenges including small area growth (up to a few μm), uncontrolled layer thickness (mono to multi-layers), randomized and non-uniform growth, and the lack of reproducibility. Due to this, large scale growth of InSe layers for electronic device fabrication has remained an area of great interest.

To overcome the aforementioned challenges, researchers started exploring alternative routes of InSe synthesis such as thermal evaporation[7], sputtering[8], chemical vapor deposition[9], molecular beam epitaxy[9] *etc*. in recent times. Although the results produced by such techniques have been promising, they lack optimization which indicates that the quest for an optimized standard growth technique for InSe films is going to be a long process.

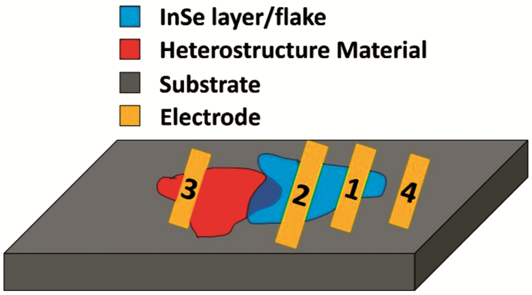

Fig. 1 — Schematic representation of an InSe heterojunction based broadband photodetector in different electrode configuration and operating mode

## 3 Device Structure

Though InSe based photodetectors can be operated in multiple configurations, a schematic representation of a typical InSe standalone as well as heterojunction based broadband photodetector is shown in Fig. 1. The diagram displays device structures with different metal electrodes (1, 2, 3 & 4) which are used to draw the output from a InSe based photodetector in diode or transistor geometry. While electrode configurations 1 & 2 are utilized to draw output from standalone InSe layer/flakes, the electrodes can be used in different configurations (1 & 3, 1 & 4, or in 3 electrode system) to draw output from InSe layer/flakes heterojunction based photodetectors.

It has been reported that InSe layer/flakes in heterojunction with other material systems such as 2D materials, metal-oxides, polymersand wide bandgap semiconductors offer optimum photon absorption, broadband photodetection, ultrahigh quantum efficiency and fast switching rate as explained in the laterpart[10-15].

## 4 InSe based Photodetectors

Research on InSe based photodetectors was initiated inthe early 2000's, but it was only in 2014 when Tamalampundi *et al.*[10] first reported the fabrication of InSe based broadband photodetector (450-785 nm) on rigid$SiO_2$/Si as well as flexible Polyethyleneterephthalate (PET) substrate. It was observed that InSe/$SiO_2$/Si based photodetector displayed a high photoresponsivity of $12 AW^{-1}$($\lambda$ = 450 nm, P = 0.66 $mWcm^{-2}$) with a switching rate of 50ms.The photodetectors fabricated on flexible substrates also revealed comparable results with or without bending of the devices. Interestingly, the observed value of responsivity *i.e.*, 12 $AW^{-1}$ was superior over other 2D materials (graphene, $MoS_2$, GaSe *etc*.), and the ease of developing flexible photodetectors[10] then ignited further research in the domain. Since then, researchers have explored various approaches to enhance and optimize the optoelectronic merits of InSe based photodetectors.

In 2015, Feng *et al.*[11] employed multilayer InSe nanosheets for broadband photodetection (UV-Vis-IR) and reported a peak photoresponsivity of $10^4$ $AW^{-1}$ surpassing the currently available silicon based photodetectors. A strong dependence of photon absorption on InSe layer thickness was perceived and the fabricated InSe phototransistors exhibited a mobility of 32 $cm^2$ $V^{-1}s^{-1}$ with significantly high current on/off ratio (~$10^7$). The study also showcased that the device performance parameters *i.e.*, photoresponsivity, detectivity *etc*. can be effectively controlled via applied gate voltage. To improve the switching rate of InSe photodetectors, Luo *et al*.[12] modified the electrode work function and replaced the metal electrode material with graphene. The InSe/Graphene photodetectors exhibited broad spectrum photodetection (400 – 1000 nm) with a response and recovery time of 120 µs and 220 µs, respectively. The obtained switching time of InSe/Graphene photodetector was 40 times faster than InSe/metal photodetector[11].

In the consecutive years, various approaches such as surface oxidation doping[13]modified chemical phase exfoliation[14], focused ion beam etching[15] *etc*. were also explored to modify the inherent material properties and alter the device structure for the optimization of photodetector's overall performance. In surface oxidation technique[13], the carrier concentration in InSe multilayers was modulated through the formation of a surface oxide layer which created a built in potential and band bending at the InSe/Oxide interface for ultrahigh gain. This led to the formation of a carrier concentration induced photo generated charge separation and resulted in a record high photoresponsivity of 5 × $10^6$ A/W. Further, extraordinary photo responsivities (> $10^7$ A/W) using InSe photodetectors were obtained via two approaches *i.e*., modifying the growth technique and changing the device fabrication technique. Kang *et al*.[14] demonstrated liquid phase exfoliation of InSe flakes in a surfactant free, low boiling point ethanol-water co solvent system. The pristine InSe flakes/films produced via this technique possessed minimal chemical residue and the fabricated photodetectors showcased a responsivity of 5 × $10^7$ A/W, which is the highest reported value till date. Comparable results (3 × $10^7$ A/W) were also produced by Yang *et al*.[15]on using the focused ion beam method to fabricate InSe/Pt photodetectors with low contact resistance *i.e*., 990 - 4400 Ωµm. Both of the reported photo responsivities were claimed to surpass and have higher magnitude than other available 2D materials such as graphene, $MoS_2$, $WS_2$, $WSe_2$, GaSe *etc*. based photodetectors[14,15].

In the meantime, research on the development of self-powered *i.e*., zero bias and heterojunction based InSe photodetectors became prevalent [16-18], but unfortunately the self-powered operation led to a significant compromise in the photoresponsivity as evident from Table 1. In early 2018, a self-powered GaTe-InSe vertical p-n heterojunction based photodetector with a responsivity of 13.8 $mAW^{-1}$ was reported by Feng *et al*.[16] The mechanism of self-powered operation was attributed to band bending and type II band alignment at the GaTe/InSe interface which creates a path for an unobstructed chargetransfer[16]. In another report on p-Se/n-InSe

Table 1 — Year wise performance of InSe based photodetectors starting from 2014

| Device Structure | Photoresponsivity ($AW^{-1}$) | Detectivity (x $10^{11}$ Jones) | Quantum Efficiency(%) | Response Time (ms) | Recovery Time (ms) | Ref. |
|---|---|---|---|---|---|---|
| $InSe/SiO_2$&InSe/PET | 12.3 | 1.07 | 1367 | 40-50 | 4000 | 10 |
| InSe Nanosheets/$SiO_2$ | 3 × $10^4$ | 20 | - | 5 | 8 | 11 |
| InSe/Graphene/$SiO_2$ | 60 | - | $10^4$ | 0.12 | 0.22 | 12 |
| Oxide/InSe/$SiO_2$ | 5 × $10^6$ | - | - | 50 | - | 13 |
| LPE-InSe/$Al_2O_3$ | 5 × $10^7$ | - | - | 0.45 | 0.09 | 14 |
| InSe/$SiO_2$ | 1.8 × $10^7$ | 11000 | - | 25000 | - | 15 |
| InSe/GaTe/$SiO_2$ | 0.013 | - | 4.3 | 0.02 | - | 16 |
| 1D-Ge/InSe/$SiO_2$ | 0.110 | 5.6 | 50 | 30 | 37 | 17 |
| Au-NPs/InSe/$SiO_2$ | 0.369 | 33.5 | 100 | 23 | 25 | 18 |
| LPE-InSe/$SiO_2$ | 274 | 54.9 | - | 15 | 64 | 2 |
| Graphene/InSe/h-BN | 1.9 × $10^4$ | 300 | $10^6$ | - | - | 19 |
| P(VDF-TrFE)/InSe/h-BN | 1.4 × $10^4$ | 163 | - | 0.6 | 1.2 | 20 |
| InSe/DDAB/BCB/$SiO_2$ | 1 × $10^6$ | 100 | 7 × $10^7$ | 16 | 16 | 4 |
| MLG-Graphene/InSe/$SiO_2$ | 1.8 × $10^5$ | - | 6.4 × $10^7$ | 22 | 108 | 21 |
| FLG/InSe/GaTe/$SiO_2$ | 63.7 | 388 | 1.9 × $10^4$ | 0.03 | 0.03 | 22 |
| $WS_2$/InSe/$SiO_2$ | 0.061 | 2.5 | 14.5 | 0.06 | 0.07 | 23 |
| ITO/InSe/$SiO_2$ | 0.06 | - | - | - | - | 5 |
| InSe/$Bi_2O_2Se$/$SiO_2$ | 13.6 | 10.8 | 4.1 × $10^3$ | 5.8 | 15 | 24 |
| $MoTe_2$/InSe/$SiO_2$ | 0.433 | 16.5 | 100 | 0.099 | 0.177 | 25 |

heterojunction based broadband photodetector, Shang et al.[17] reported a high on/off ratio of $10^3$ with a photoresponsivity of 110 $mAW^{-1}$ under self-powered mode. The type II alignment at the p-Se/n-InSe interface was ascribed as the reason for self-powered operation of the photodetector. Thereafter, researchers used InSe layers and Au nanoparticles array to fabricate self-powered dual band photodetectors[18]. InSe nanosheets in combination with the surface plasmon resonance of Au nanoparticles improved the photocurrent and increased the responsivity to 369 $mAW^{-1}$.

In InSe based phototransistors, doping of InSe and modification in metal and gate electrode material have shown promising results[12-14,19,20]. Wang et al.[4] carried out molecular functionalization of InSe with surfactant didodecyldimethylammonium bromide (DDAB) which resulted in broadband photodetection with ultrahigh mobility ($>10^3$ $cm^2V^{-1}s^{-1}$) and photoresponsivity ($>10^6$ $AW^{-1}$). The functionalization has shown to increase the doping density ($> 10^{12}$ $cm^{-2}$), lowering the fermi level and healing of defects states in InSe for high photoresponsivity ($10^6$ $AW^{-1}$) and fast switching (5 ms). It was observed that changing the metal gate electrodes with graphene[19] and P(VDF-TrFE)[20] has shown significant improvement in device's optoelectronic merits. InSe photodetectors with vertical few layers graphene (FLG) contacts have shown remarkable performance with 12 times more responsivity over photodetectors with metal contacts[19]. On the other hand, P(VDF-TrFE) copolymer gated phototransistors displayed very high on/off ratio ($\sim 10^8$) due to the suppression of dark current[20].

The fabrication of InSe heterojunction based photodetectors has been very prominent in the last 2-3 years. Various material systems such as graphene[21], MMCs[22], TMDCs[23,24], Oxides[5,25] *etc.* were explored and utilized to develop InSe based efficient and broadband photodetectors. The complimentary properties of these materials assist in optimizing the performance of InSe photodetectors.

Cui *et al.*[21] fabricated InSe and multilayer graphene (MLG) *i.e.*, MLG/InSe heterostructure based photodetectors and observed that built-in electric field at MLG/InSe interface and the photogating effect leads to significant enhancement in photon absorption.The responsivity ofMLG/InSe photodetector also divulged five orders of magnitude higher than that of sole InSe photodetector. In InSe/GaTe Van der Waal heterojunction[22] based photodetector, the built-in electric field associated elimination of series resistance and confinement of charge carriers in the depletion region displayed excellent optoelectronic characteristics with a responsivity of 63.7 $AW^{-1}$ and response time of 30 µs. Using $WS_2$, Chen *et al.*[23] demonstrated that $WS_2$/InSe based n-n photodetector got similar results *i.e.*, responsivity = 61 $AW^{-1}$, with large on/off ratio ($10^8$) and response/decay time of 63/76 µs. The reason behind these results was also attributed to the inbuilt electric field[23]. Recently in 2024, He *et al.*[24] utilized MoTe2/InSe Van der Waal heterostructure to fabricate a self-powered broadband photodetector and produced a comparatively better responsivity of 433.88 $mAW^{-1}$ with a significantly fast response/decay time of 99/117 µs in comparison with previously reported self-powered photodetectors[17-19].

Literature has very limited notable studies on non van der Waals2D materials based InSe photodetectors due to their poor performance. Zhang et al.[25] reported $Bi_2O_2Se$ Nanosheet/InSe Nanoflake based Type II Homo type self-driven broadband photodetectors with a responsivity of 13.3 $mAW^{-1}$. Despite a large $I_{light}/I_{dark}$ ratio of $10^5$, the device had an average switching rate (5.8/15 ms). All the aforementioned studies not only attempt to enhance the device performance but also improve their ambient stability[5]. It is expected that such approaches altogether will contribute significantly to the advancement of InSe based optoelectronic device technology.

## 5 Challenges and Future Aspects

Despite the significant advancements in InSe based photodetectors in recent years, the technology is still far from mature and suffers numerous challenges. The first major challenge is large area ($>100$ $\mu m^2$), uniform and optimized growth of InSe thin films. Till now, the widely used synthesis process of InSeis mechanical/chemical exfoliation which produces small ($<10$ µm) and randomly oriented InSe flakes.InSe thin films/flakes produced by these techniques cannot be optimized or reproduced. The second major challenge is control over layer thickness (mono to multi-layer). Due to van der Waals interaction, the electronic, optical as well as structural properties of InSe are highly dependent on InSe layer thickness which govern the device output. Other notable challenges include phase transition (InSe to $In_2Se_3$), air stability (*i.e*., ambient oxidation) and transfer of the synthesized InSe thin films on intended substrates to synthesize the desired heterostructures.

As InSe pursues tunable bandgap, excellent mobility and lower value of young modulus (upto 23.1 GPa) among all crystalline two dimensional materials, it can sustain large deformations which makes it an excellent candidate for ultrathin and flexible optoelectronic devices operating in a broad spectral of electromagnetic radiation. Also, besides optoelectronics, InSe has profound applications in photonics (photovoltaics), sensing (environmental monitoring) and imaging (optical tweezers) technology.

## 6 Conclusion

In this mini review, we have highlighted some of the notable recent developments in the field of InSe based photodetectors. We have outlined the importance of InSe based photodetectors in optoelectronics. The review briefly explains the growth techniques, their merits and architecture of anInSe based photodetector in diode and transistor configuration. We have discussed about self-powered photodetectors using standalone InSe films and in heterojunction with other material systems. It was observed that the best reported values of responsivity, detectivity, response and recovery time for an InSe based photodetectors are $5 \times 10^7$ $AW^{-1}$, $1.1 \times 10^{15}$ Jones, 32 μs and 34 μs, respectively. Recent developments suggest that InSe based heterostructures can lead to the fabrication of broadband photodetectors, though challenges related to large area growth, layer thickness and air stability needs to be addressed.

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