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Manipulating Optical Absorption of Indium Selenide Using Plasmonic Nanoparticles

Xiaoyu Liu, Yifei Zhang, Huayu Feng, Yafei Ning, Yanpeng Shi,* Xiaodong Wang, and Fuhua Yang

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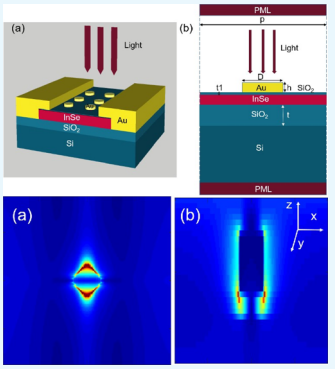
ABSTRACT: In this work, we propose using periodic Au nanoparticles (NPs) in indium selenide-based optoelectronic devices to tune the optical absorption of indium selenide. Electromagnetic simulations show that optical absorption of indium selenide can be manipulated by tuning plasmonic resonance. The effect on the plasmonic resonance of the size, period of NPs, the thickness of silicon oxide, and the insulator spacer is systematically analyzed. A high absorption enhancement over the visible spectrum is achieved through systematic optimization of nanostructures.



1. INTRODUCTION

After the successful exfoliation of graphene, two-dimensional (2D) layered materials have attracted huge attention in the past decades, which exhibits excellent carrier transport because of its unique 2D energy dispersion.[1−3] Graphene shows high responsivity in photodetectors and photovoltaics; however, the dark current is high as a result of the disappearance of the band gap.[4−6] In addition, transition-metal dichalcogenides also exhibit novel optical properties such as MoS_2.[7] However, the band gap of MoS_2 is 1.8 eV, which limits its application in photovoltaic solar cells and near-infrared photodetectors. Indium selenide (InSe), one of the members in the family of the layered metal chalcogenide semiconductors, is considered to be a potential 2D material for the future Si-based optoelectronic devices.[8−11] InSe has its unique characteristics. Compared with MoS_2, the direct band gap of bulk InSe is narrower ($E_g \approx 1.3$ eV). Nevertheless, InSe covers the solar spectrum well, providing a wider spectral response.[12] From the monolayer to the bulk, the band gaps show a dramatic change because of the strong interlayer interaction of InSe.[13] The band gap of the InSe bulk material is stable and moderate, and layered InSe undergoes a band gap transition process from indirect to direct, which makes it have extraordinary application prospects in the field of photoelectric detection.[14] In addition, the InSe ridge offers a new platform for photovoltaic applications or wide band spectrum photodetectors, making tunable optoelectronic devices overlap well in the near-infrared regime. A band gap tuning window as large as ∼1.1 eV in 2D InSe has been predicted by first principles calculations, and experimentally, it has been proved that in 5 nm thick InSe nanosheets, the optical band gap exhibits a 0.2 eV blue shift.[15] InSe becomes a competitive material for the applications in electronics and optoelectronics because of the high carrier mobility, ambient stability, and layer-tunable band gap.[16−20]

Generally, optoelectronic devices with high performance need two factors, electron mobility and efficient light absorption.[21−24] On the one hand, high electron mobility is desired to separate the electron−hole pairs in time. It has been proposed that the multilayer InSe field-effect transistors on the SiO_2/Si substrate can obtain a high mobility of 162 $cm^2\ V^{-1}\ s^{-1}$ at a certain thickness.[25] By utilizing poly(methyl methacrylate)/Al_2O_3 as the dielectric layer instead of SiO_2, the electron mobility can be further increased up to 1055 $cm^2\ V^{-1}\ s^{-1}$ because carrier scattering is significantly reduced by polar phonon scattering and surface-charged impurities.[26] On the other hand, efficient absorption should be obtained to generate enough electron−hole pairs. However, the optical absorption of InSe with a limited thickness of 30 nm is intrinsically poor, which results in low photoresponsivity. For the electric field polarized perpendicular to the layer plane, the dipolar selection rules of 2D InSe favor optical transitions to occur, which leads to a powerful in-plane/out-of-plane anisotropy of the optical properties.[15] Under normal incidence

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of a plane wave, the optical absorption of InSe is low for electric fields with in-plane polarization, especially with limited thickness. A way to increase the optical absorption is to combine plasmonic nanostructures based on noble metals with InSe, and the responsivity of the visible spectrum is tunable through utilizing the localized surface plasmon resonance (LSPR) of metal nanoparticles (NPs).[27] Plasmonic nanoantennas based on noble NPs can promote the performance of InSe-based optoelectronic devices because of near-field excitation and hot electron contribution. Photovoltage and photocurrent in InSe-based devices display powerful responsivity enhancement with the application of NP arrays.[28] Despite initial experimental studies, it should be noted that systematic modeling of the nanostructure-based InSe photodetectors has not been studied at present, to achieve high absorption enhancement and spectral selectivity.

In this paper, we systematically reveal the plasmonic effects of gold NP array nanostructures on the InSe layer. The light absorption of the InSe layer is manipulated by tuning the plasmonic resonance of Au nanoantennas. Our research demonstrates that plasmonic resonance can be controlled to generate alternative optical absorption enhancement, originating from the light concentration of LSPR and the electromagnetic coupling between gold nanoantennas and InSe.

2. RESULTS AND DISCUSSION

As shown in Figure 1, the periodic gold NPs are cylinders with a diameter of D and height h, which are attached to InSe on a

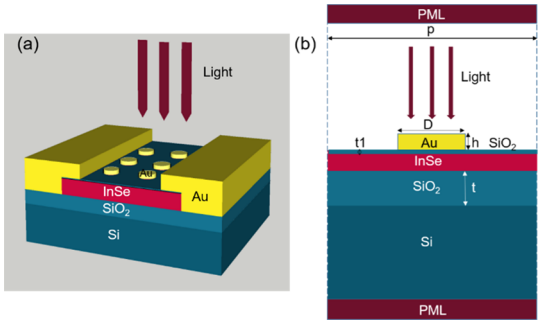

Figure 1. (a) Schematic diagram and (b) cross-sectional view of the proposed InSe-based optoelectronic device.

SiO_2/Si substrate. The refractive index of silicon oxide is n = 1.46 and the thickness of silicon oxide, the period of NP arrays and the thickness of the inserted silicon oxide are defined as t, p, and t_1, respectively. The optical constants of simulated gold NPs, InSe, and silicon in the structure are taken from the obtained experimental values proposed in the literatures,[29,30] and the out-of-plane absorption of InSe is not taken into consideration, which means that InSe is assumed to be isotropic, so as all other refractive media. Here, the influences of heat disturbance and saturable absorption in InSe are ignored. The finite-difference-time-domain approach is used with perfectly matched layers as the optical open boundary conditions to explore the optical absorption of InSe by electromagnetic simulation. The discretization of space adopts 0.05 times of the metallic structure's size in x, y, and z directions, which was verified to offer convergence of the simulated results with no loss in physical detail. We only consider the normally incident plane electromagnetic wave with x-polarization.

2.1. Size and Period of Au NPs. To accurately estimate the absorption enhancement, we first simulate the optical absorption of InSe flakes on SiO_2/Si as a reference, as shown in Figure 2a. It can be found that the absorption of InSe is quite

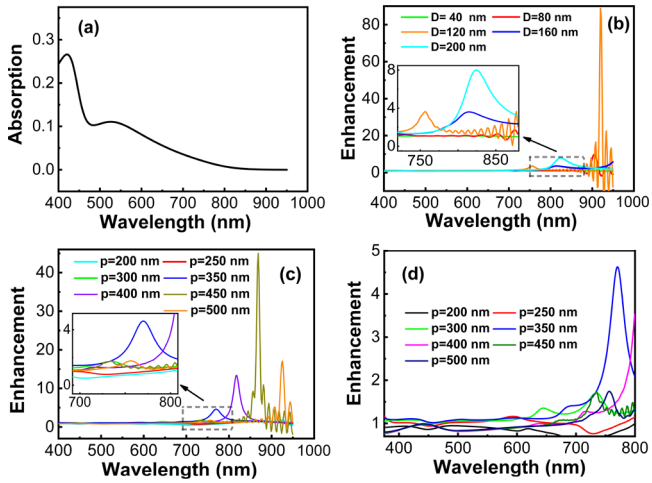

Figure 2. Absorption spectra of InSe with a thickness of 30 nm on silicon (a) without Au NPs, (b) with the diameter of Au NPs varying, p fixed at 500 nm, (c,d) with the period of Au NPs varying, D fixed at 120 nm.

low from 600 to 950 nm, less than 10%, while the absorption of 400−600 nm is relatively higher. As seen in Figure 2b−d, the optical absorption enhancement spectra cover the entire spectral range, with different physical mechanisms in different bands. The localized surface plasmons of the metal act on the optical absorption in the long wavelengths, with the electric field localized in the InSe layer, thereby increasing optical absorption. Also, in the 400−700 nm wavelength region, the reflectance of the surface is reduced because of the Mie scattering effect of the metal NPs, thus increasing the absorption of InSe. In this regard, we investigate plasmonic resonance for absorption enhancement of InSe, particularly in the long wavelengths. Substrate effects, particle size, environmental dielectric media, and electromagnetic coupling between periodic particles can greatly influence the plasmonic resonance of gold NP arrays. The electron plasma oscillations (particle plasmons) in single metal NPs have great impact on the optical extinction spectrum in the visible range. The light absorption of InSe can be governed by the above factors through tuning the plasmonic resonance. First, the influences of the array period p and diameter D on the optical absorption enhancement in the InSe layer are investigated. Meaningful variations of the width and position of the particle-plasmon resonance were demonstrated, which could be attributed to in-phase superposition of scattered light from neighboring particles.[31] When NP sizes and array periods are altered, the frequency of the plasmonic resonance peak could be manipulated, thus tuning the optical absorption of InSe, as shown in Figure 2b−d. Despite tuning the plasmonic resonance, NPs also can enhance optical absorption as optical scatters. According to Mie scattering theory, the scattering efficiency is high when the size of the scatter is on the order of the light wavelength. Hence, p is fixed at 500 nm as shown in Figure 2b, and the diameter D is changed from 40 to 200 nm. As the absorption is quite low in the long wavelengths, the absorption enhancement spectra, with respect to the

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absorption of InSe without Au NPs are shown in Figure 2b. It reveals that the enhancement is high, up to nearly 90. The enhancement peak value in the long wavelength has a redshift as the period p increases, which is attributed to the coupling between localized plasmons. The diameter D = 120 nm is selected in the following simulations as the absorption enhancement is high over most of the visible spectrum. Then the period of Au NPs is studied. According to Mie scattering theory, the electromagnetic interactions between Au nanoantennas can be ignored and the absorption in InSe presents weak enhancement if the NP period is much bigger than NP size, which can be explained by the fact that the strong near field in InSe is highly concentrated surrounding the bottom edges of Au NPs, shown in Figure 2c. When the fill factor becomes larger, electromagnetic coupling between NPs enhances the intensity of the optical near field in the part of InSe where no Au NPs are located. Until the maximum spectrum peak at the optimum particle density is reached, the absorption capability of InSe has been markedly improved, which is displayed in Figure 2d. In the short wavelength, the enhancement increases when p varies from 200 to 350 nm and decreases when p varies from 350 to 500 nm. Therefore, the diameter D and period p are set to 120 and 350 nm in the following calculations.

2.2. Underlying Physics of the Optical Absorption Enhancement. This underlying physics can be intuitively elaborated by the electric field diagram shown in Figure 3,

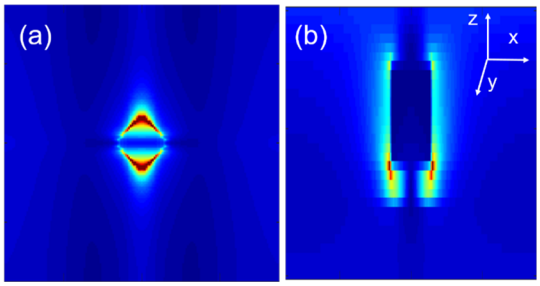

Figure 3. Electric field intensity distribution diagram at wavelength 769 nm, (a) $x-y$ axis view (b) $x-z$ axis view.

which demonstrates that absorption enhancement is attributed to the influence of plasmonic light concentration by using gold NPs. As shown in Figure 3a, light energy is concentrated around the bottom edge of the cylinder. The light source is polarized, resulting in the two semicircle-like pictures as shown in Figure 3a. The concentrated energy would be a whole circle with natural light. As shown in Figure 3b, the forward scattering is much stronger than the back scattering, which means that most light energy at 769 nm is scattered into the layer of InSe. Because the SiO_2 layer is very thin, the optical near field is enhanced and concentrated in the InSe layer, and InSe has an absorption peak induced by LSPR at this wavelength. When the wavelength of the scattered light is appropriate to period p, a geometric resonance appears, which could cause a dramatic modification of the measured optical extinction when it happened at the same wavelength as the LSPR.

2.3. Thickness of Silicon Oxide and the Height of Au NPs. Then, the influence of the silicon oxide dielectric layer is researched. The thickness of silicon oxide, which is located between the active layer and reflective substrate Si, can be adjusted to tune the optical absorption of the active layer.[32] As shown in Figure 4, we investigate the effect of silicon oxide thickness on optical absorption enhancement of the InSe layer. Because of the phase delay of the reflective wave from InSe compared to the reflective wave from Si, the InSe layer shows interferometric absorption on a SiO_2/Si substrate without Au NPs. Because of the interference effects, the maximum reflection and minimum absorption appears periodically. The effects of both the plasmonic resonance and optical interference have to be considered for the design of nanostructures, to achieve optimal enhanced absorption using Au NPs. When the intrinsic interferometric absorption peak is suited to the plasmonic resonance wavelength, the enhanced near field efficiently promotes the optical absorption of InSe. Figure 4a displays the absorption of InSe with varying thicknesses of SiO_2, and the absorption enhancement spectra compared to the absorption spectrum of InSe with 100 nm SiO_2 are displayed in Figure 4b. Plasmonic resonance of gold NPs gives rise to the maximum absorption at t = 300 nm and the intrinsic absorption peak occurs because of optical interference, and they are perfectly matched at 769 nm. The thickness of silicon oxide slightly affects the position of the plasmonic resonance, from 769 to 779 nm. Meanwhile, the coupling between the optical interference and plasmonic resonance plays an important role in the enhancement of the light absorption, from 1.22 to 4.78, which means that the light absorption enhancement is weakened by the mismatch between plasmonic resonance and optical interference. To

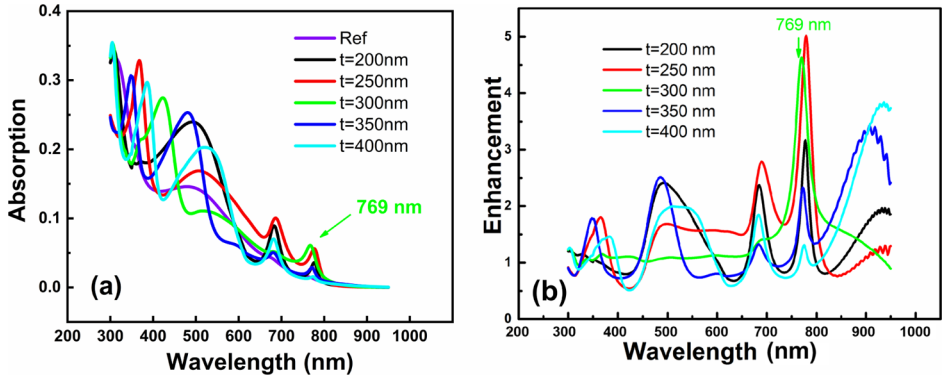

Figure 4. (a) Absorption ratio and (b) absorption enhancement spectra of InSe with the varying thickness of SiO_2 with NPs (SiO_2 of 100 nm as a reference).

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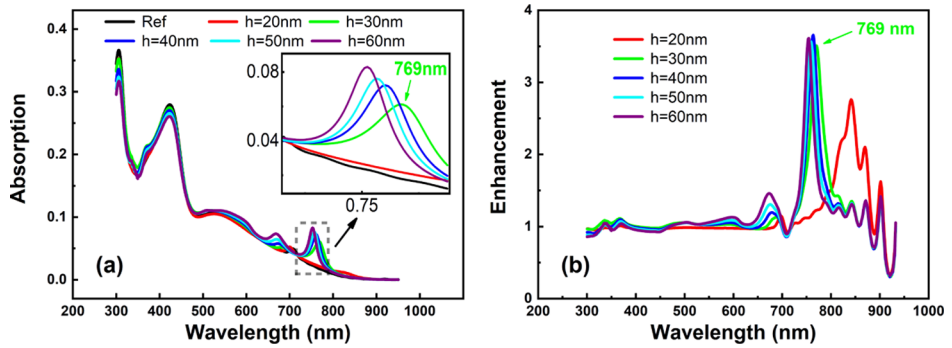

Figure 5. (a) Absorption ratio and (b) absorption enhancement spectra of InSe with different heights of Au NPs (Au NPs of 10 nm as a reference).

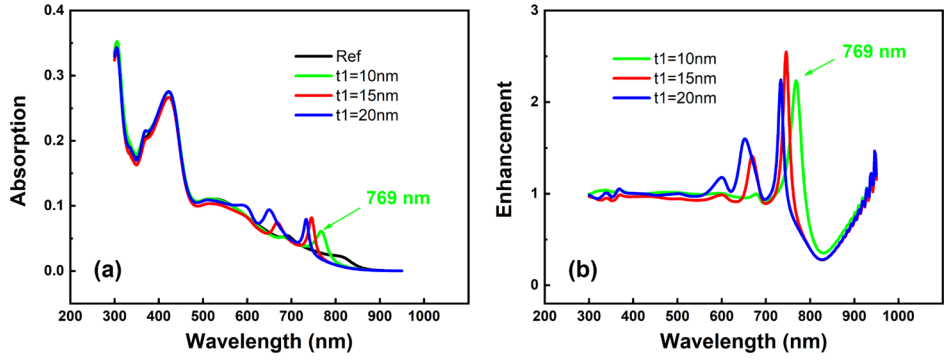

Figure 6. (a) Absorption ratio and (b) absorption enhancement spectra of InSe with different inserted SiO_2 thicknesses (Inserted SiO_2 of 5 nm as a reference).

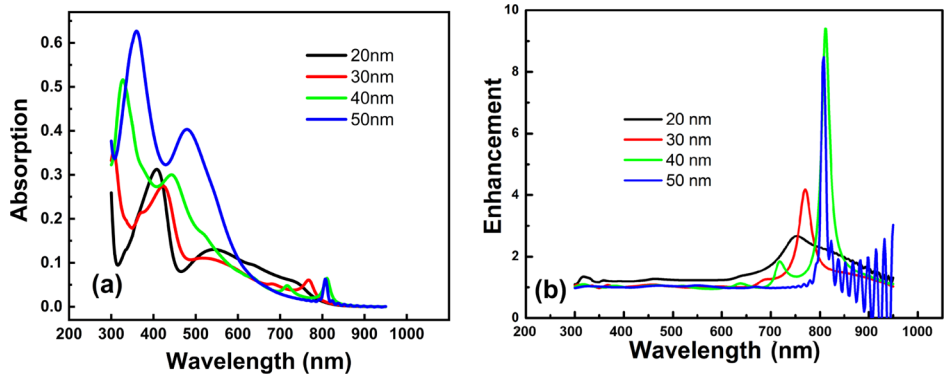

Figure 7. Au NPs on (a) absorption ratio and (b) absorption enhancement spectra of InSe with different thicknesses.

facilitate further research, we use a thickness of 300 nm for silicon oxide in the following simulations.

Next, we investigate the influence of Au particle height *h* on the absorption of the InSe layer. As shown in Figure 5a,b, the absorption spectra showed dramatic enhancement on account of plasmonic resonance because of Au NPs. When *h* increases from 10 to 60 nm, the enhancement peak value displays a blue shift as shown in Figure 5a, which is attributed to the change of plasmonic resonance wavelength by manipulating *h*. When *h* is small, such as smaller than 30 nm, the plasmonic resonance is closely related with *h*. When *h* equals to 10 nm, the Au NPs can be seen as a nanodisk, while when *h* equals to 30 nm, the Au NPs can be seen as a cylinder. The near-field effects of LSPR mainly concentrated on the lower surface of gold NPs. Therefore, when *h* is larger than 30 nm, it has little impact on near-field effects and the blue shift of the plasmonic resonance wavelength becomes small which is consistent with Figure 5b.

2.4. Insertion of Insulators. For real device applications, the quantum efficiency of optoelectronic devices may be significantly lowered by carrier transport and exaction recombination on the interface between InSe and Au NPs. Hence, the isolation between InSe and gold NPs is necessary. As shown in Figure 6a,b, the influence of using SiO_2 spacer as a transparent insulator spacer has been investigated. Figure 6a displays that the insulator has a similar effect at the short wavelengths from 300 to 600 nm. The absorption peak results from the excitation of LSPR in the long wavelengths from 700 to 900 nm. The LSPR varies with the changes of the surrounding environment. Thus, as t_1 increases, the LSPR displays a blue shift. As seen in Figure 6b, the absorption enhancement is slightly weakened by increasing the thickness of the spacers as near-field effects of LSPR are weakened.

2.5. Absorption of InSe with Different Thicknesses. To verify the application of the Au NPs on InSe with different

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thicknesses, the absorption of InSe with different thicknesses are simulated, as shown in Figure 7a,b. For InSe with a thickness of 20 nm, Au NPs enhance the absorption over the whole spectrum, which can be used in optical devices such as photovoltaic devices and broadband photodetectors. With the thickness increases, the enhancement value goes down as the absorption of InSe without Au NPs increases. However, the enhancement effect becomes obvious at the long wavelengths, which is meaningful to narrow band detectors as shown in Figure 7b. Therefore, for different applications, one can manipulate the absorption in the InSe layer through designing the Au NPs. The enhancement value oscillates due to optical interference effect when the thickness is larger than 50 nm.

3. CONCLUSIONS

In conclusion, we demonstrate that the InSe-based optoelectronic devices can be combined with periodic Au NPs to promote optical absorption enhancement and alternation. The optical absorption can be tuned by plasmonic resonance of Au nanoantennas through changing the thickness of silicon oxide, the insulator spacers, and the size and period of Au NPs. The outstanding absorption efficiency and selectivity open up a channel to increase the light responsibility of InSe by plasmonic near-field effects and show great potential in optoelectronic devices, such as InSe-based photodetectors, sensors, and photovoltaics.

AUTHOR INFORMATION

Corresponding Author

Yanpeng Shi − *Center of Nanoelectronics and School of Microelectronics, Shandong University, Jinan 250100, China;* orcid.org/0000-0003-1970-5783; Email: ypshi@sdu.edu.cn

Authors

Xiaoyu Liu − *Center of Nanoelectronics and School of Microelectronics, Shandong University, Jinan 250100, China*

Yifei Zhang − *Center of Nanoelectronics and School of Microelectronics, Shandong University, Jinan 250100, China;* orcid.org/0000-0001-6077-8525

Huayu Feng − *Center of Nanoelectronics and School of Microelectronics, Shandong University, Jinan 250100, China*

Yafei Ning − *Center of Nanoelectronics and School of Microelectronics, Shandong University, Jinan 250100, China*

Xiaodong Wang − *Engineering Research Center for Semiconductor Integrated Technology, Institute of Semiconductors, Chinese Academy of Sciences, Beijing 100083, China*

Fuhua Yang − *Engineering Research Center for Semiconductor Integrated Technology, Institute of Semiconductors, Chinese Academy of Sciences, Beijing 100083, China*

Complete contact information is available at: https://pubs.acs.org/10.1021/acsomega.9b03949

Notes

The authors declare no competing financial interest.

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ABBREVIATIONS

NP, nanoparticle; 2D, two-dimensional; InSe, indium selenide; LSPR, localized surface plasmon resonance

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