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Chapter

Simple Preparations for Plasmon-Enhanced Photodetectors

Yu Liu, Junxiong Guo, Jianfeng Jiang, Wenjie Chen, Linyuan Zhao, Weijun Chen, Renrong Liang and Jun Xu

Abstract

Localized surface plasmon resonance (LSPR), known as the collective oscillation of electrons and incident light in metallic nanostructures, has been applied in high performance photodetectors over the past few years. But the preparation process is complex and expensive due to the introduction of electron beam lithography (EBL) for preparing nanostructures. In the past few months, we have demonstrated two simple methods to prepare plasmon-enhanced photodetectors: (i) Au nanoparticles (Au NPs) solution were directly spun coated onto the WS$_2$-based photodetectors. The performance has been enhanced by the LSPR of Au NPs, and reached an excellent high responsivity of 1050 A/W at the wavelength of 590 nm. (ii) Au NPs were deposited on MoS$_2$ by magnetron sputtering. The spectral response of pure MoS$_2$ was located in visible light and which was extended to near-infrared region (700–1600 nm) by Au NPs. Further, the responsivity reaches up to 64 mA/W when the incident light is 980 nm. In this book chapter, more details for developing those two simple methods and the discussion of the enhanced mechanism are performed, which can be very useful for the next generation photodetection.

Keywords: surface plasmon resonance, Au nanoparticles, enhanced photodetectors, simple preparations, high responsivity

1. Introduction

Photodetectors are one of the most important devices in photonic chips, which show a great potential in optical communication, flame sensing, environmental monitoring, and astronomical studies [1]. A lot of semiconductors have been exploited for photodetection such as silicon, GaN, PbS, InGaAs and HgCdTe, operating from ultraviolet to far-infrared region [2]. It is significant to decrease the dimension of photodetectors down to the sub-nano scale for the next generation highly integrated photonic chips.

Two-dimensional (2D) materials such as graphene and layered transition metal dichalcogenides (TMDs) have attractive electronic and optical properties such as flexibility, transparency and metal-oxide-semiconductor (CMOS) compatibility [3–7]. Tungsten disulfide (WS$_2$), a typical member of the TMDs group, has a higher carrier mobility than other TMD materials due to smaller electron effective mass [8]. Moreover, WS$_2$ enjoys excellent thermal stability for extensive applications [9].
Hence, WS$_2$-based high stable photodetectors can be used in many attractive applications such as extreme environment detection. Furthermore, Two-dimensional stacked molybdenum disulfide (MoS$_2$) has attracted many research interests for applications in optoelectronic devices, due to its outstanding merits of electronic and optical properties, especially photodetectors. But the few layers 2D material-based photodetectors suffer from low photoresponsivity mainly due to the poor optical absorption in the atomic layered materials.

A number of 2D material-based photodetectors have been enhanced using resonance microcavity, PbS quantum dots, tunneling effect, heterojunctions or perovskite [10–14]. The narrow band absorption or complex preparation process limits the application of these attractive methods. Recently, localized surface plasmon resonance (LSPR)-enhanced photodetector has been demonstrated, and we have also provided effective ways to enhance the efficiency of light-harvesting [15–17]. LSPR can be excited by Ag or Au nanoparticles (NPs) such as deep metallic grating, nanodisk array, bowtie array, hybrid antenna and fractal metasurface [18–20]. Furthermore, patterning 2D materials into periodic structure can also excite plasmon resonance [21]. The methods for preparing the nanostructure involve electron beam lithography, hydrothermal synthesis, and template-based electrochemical method, which are generally complicated, expensive and may deleterious to the device.

For the current existing problems, we have demonstrated two methods for easy-preparation and high-performance 2D material-based photodetectors [22, 23]. (i) Au NPs solution was directly spun coated onto WS$_2$-based photodetectors. The performance has been enhanced by the LSPR of Au NPs, and reach quite high responsivity of 1050 A/W at the wavelength of 590 nm. The diameters and distance of Au NPs will affect the resonant wavelength and absorption of the device. (ii) We have demonstrated a MoS$_2$ plasmonic photodetector by depositing Au NPs on MoS$_2$ sheet using magnetron sputtering without need of template, which shows a significant improvement of photo-response in near-infrared region. The spectral response of pure MoS$_2$ was in visible light and which was extended to near-infrared region (700–1600 nm). Furthermore, the responsivity reaches up to 64 mA/W when the incident light is 980 nm. Detailed preparation and discussion of the mechanism are performed in the chapter.

2. The preparation and theoretical mechanism of spun-coated WS$_2$-based photodetectors

2.1 The preparation process

The WS$_2$ film was grown on sapphire substrate by chemical vapor deposition (CVD) method and transferred to Si/SiO$_2$ substrate by wet transfer method. The molecular configurations of WS$_2$ are shown in Figure 1a. Raman spectra (Figure 1b), PL spectrum (Figure 1c) and atomic force microscopy (AFM) files (Figure 1d) reveal monolayer feature of WS$_2$ film.

The 3D and cross-section view of the photodetector is shown in Figure 2a, b, respectively. The light incident normally from the top of the device. The fabrication was carried out by the following steps. First of all, a 200-nm-thick molybdenum (Mo) layer was deposited on WS$_2$ layer. Afterwards, the WS$_2$ was patterned (size: 30 × 100 μm) by photolithography and plasma etching to form active area. Finally, Au NPs solution was spun coating onto the channel and dried in air. Nearly spherical Au NPs can be seen in the low-resolution transmission electron microscopy (TEM) image (Figure 2c). The mean diameter of Au NPs is ~20 nm.
Figure 1.
The microscopic molecular structures and characterization of monolayer WS$_2$ film [22]. (a) Schematic molecular structure of 1 L-WS$_2$. The blue and yellow balls present sulfur and tungsten, respectively. (b) The Raman spectrum consisted of several characteristic peaks of the WS$_2$ film on Si/SiO$_2$ substrate acquired with laser excitation of $\lambda=532$ nm. (c) PL spectra of 1 L WS$_2$ layer. The band gap is about 1.96 eV as shown in the inset. (d) The AFM height profiles and corresponding AFM image of 1 L-WS$_2$. The thickness is about 0.8 nm.

(Figure 2d) as shown in the statistical analysis of the TEM images. Typical low-resolution (Figure 2e) and high-resolution (Figure 2f) scanning electron microscope (SEM) images of the Au NPs are also presented. The Au NPs are well distributed on the top of the WS$_2$ film.

2.2 The performance of the WS$_2$-based photodetector

The drain-source current ($I_{DS}$) under illumination at room temperature without Au NPs are shown in Figure 3a. It is concluded that $I_{DS}$ increase as $V_{DS}$ increase from 0 to 2 V. The irradiance power is 20.5 mW/cm$^2$ in all of these three wavelength (590, 740 and 850 nm). $I_{DS}$ decrease with the increase of wavelength. The ratio of the on/off current ($I_{DS}/I_{dark}$) reached nearly $10^3$ under 590 nm light illumination. The responsivity can be calculated by

$$R = \frac{I_{Ph}}{P}$$  (1)

where $I_{Ph}$ is the photocurrent, $P$ is the irradiance power. The responsivity at $V_{DS} = 2$ V is illustrated in Figure 3b. The responsivity decrease with the increase of the power which are typical for photodetectors [24, 25]. $R$ reached 35 A/W at the wavelength of 590 nm, and reached 1.8 A/W at the wavelength of 850 nm when irradiance power is both 0.2 mW/cm$^2$. The performance of the photodetector decoated with Au NPs is shown in Figure 3c. The drain-source current are measured at the irradiance power of 20.5 mW/cm$^2$. The enhanced $I_{DS}$ also reached the highest value at 590 nm and decreased with the increase of the wavelength ($\lambda$).

The current gain is defined as

$$G = \frac{I_{pe}}{I_{ph}}$$  (2)
where $I_{pe}$ is the enhanced photocurrent of the photodetector. The current gain reveals the improvement of the device by Au NPs as shown in Figure 3d when $P = 0.2\, \text{mW/cm}^2$ and $V_{DS} = 2\, \text{V}$. The photoresponsivity was enhanced ~30 times and reached 1050 A/W when $\lambda = 590\, \text{nm}$. The photoresponsivity was enhanced ~11 times and reached 55 A/W when $\lambda = 740\, \text{nm}$. And the photoresponsivity was enhanced ~5 times at near infrared light ($\lambda = 850\, \text{nm}$) and reached 8 A/W. In general, the switching behaviour, which reflect the response speed and high-frequency characteristic, is very important for photodetectors. The detectors also need to quickly refresh in some applications such as instant display. Figure 3e presents the switching behavior at near infrared light ($\lambda = 850\, \text{nm}$) of the WS$_2$ photodetector. The photodetector shows a good repeatability during on-off cycles. Moreover, the on-off characteristic in a period is shown in Figure 3f. The rise and decay time are about 100 and 200 ms, respectively.

2.3 The theoretical mechanism

To explain the mechanism of the enhancement by Au NPs, we take finite-difference time-domain (FDTD) method to investigate the distribution of electric field of Au NPs. According to the Förster’s expression for energy $W$ transferred from donor to acceptor [24, 25].

Figure 2. Characterization of the fabricated photodetector [22]. (a) The schematic 3D view of a L-WS$_2$-based photodetector was presented. The drain/source electrodes are fabricated by Mo. Au NPs (red balls) were spun coated on the channel. (b) The cross-section view of the photodetector. (c) TEM image of Au NPs. The Au NPs are well distributed in the solution. (d) The statistics size distribution of Au NPs based on the TEM image. We can conclude that the mean size of Au NPs is ~20 nm. (e) Low-resolution and (f) high-resolution SEM images of the photodetector. Clear electrodes and Au NPs can be seen from these images.
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DOI: http://dx.doi.org/10.5772/intechopen.89251

\[ W_d = \frac{9}{8\pi} \int_0 f_d(\omega) \sigma_a(\omega) |D|^2 \] (3)

where \( W_d \) is the donor’s energy, \( f_d(\omega) \) and \( k \) are the spectral function and wave vector of the source, \( \sigma_a(\omega) \) is the absorption of acceptor, \( D = q/r^3 \) is the coupling coefficient, and \( r \) is the distance. The performance could be changed by key parameters of Au NPs such as diameter \( d \) and distance between two particles \( s \). In order to give an intuitive description, simplified models were built. The distance between the edges of two adjacent nanospheres, \( s \), was fixed on 10 nm. The diameter \( d \) was fixed as 20 nm.

The resonant wavelength can be acquired by SPPs dispersion equation.

\[ \beta = k_0 \sqrt{\frac{\varepsilon_d + \varepsilon_m}{\varepsilon_d \varepsilon_m}} \] (4)

where \( k_0 \) is the vacuum wavevector, \( \varepsilon_d \) is the relative permittivity of dielectric, \( \varepsilon_m \) is the dielectric function of gold, which can be represented by Drude model. The mismatch of SPPs and incident wavevector can be compensated by the gold nanoparticles, which can be approximately presented by

\[ \beta = \sqrt{\varepsilon_d} k_0 \sin \theta + \frac{2\pi n}{\lambda_g} \left( \frac{x + y}{\lambda_g} \right) \] (5)

where \( \theta, \lambda_g, n \) are the horizontal angle of incident wave vector, the grating period, and an integer, respectively. From Eq. (5), we can see that the absorption wavelength depends only on the dimension of Au NPs.

The results for the illumination at \( \lambda = 590, 740, \) and 850 nm are shown in Figures 4a–c, respectively. It is clear that the intense electromagnetic fields were introduced by LSPR of Au NPs. The electric field near the Au NPs was significantly enhanced and stronger than the rest region, revealing that electromagnetic energy was compactly confined by the Au NPs. The electromagnetic field was enhanced more significant when the illumination was under \( \lambda = 590 \) nm. The
enhanced electric fields could excite the generation of the carriers in the WS\(_2\) film, resulting a prominent photoresponse. The highest responsivity obtained at \(\lambda = 590\) nm (Figure 3b, d) is consistent with the most intense LSPR at \(\lambda = 590\) nm (Figure 4a). The generation and transportation of the electrons are shown in Figure 4d. The photons were absorbed by WS\(_2\) film and the excited electrons were driven by the drain-source voltage. There are more electrons around the Au NPs as Figure 4d shows.

3. The preparation and theoretical mechanism of magnetron-sputtering-based MoS\(_2\) photodetectors

3.1 The preparation process

Figure 5a, b shows the schematic and optical image of our designed MoS\(_2\) plasmonic photodetector by introducing Au NPs, respectively. The few-layered MoS\(_2\) sheet was obtained using mechanical exfoliation method. Then, we transferred the exfoliated MoS\(_2\) to the SiO\(_2\)/Si substrate which contacted with Au/Ni electrodes. Next, we fabricated the Au NPs on exfoliated MoS\(_2\) sheet using magnetron sputtering technique. This facile method offers the convenience of without need of template compared with other reported MoS\(_2\) plasmonic photodetectors.

Figure 6 shows the morphology of as-prepared materials was characterized using an AFM. As shown in Section A1 in Figure 6a, b, it indicates the thickness of
exfoliated bare MoS$_2$ is just about 6 nm (about 10 folds). In comparison, the surface morphology of the MoS$_2$ after depositing with Au NPs by magnetron sputtering was shown in Figure 6c–f. It clearly exhibits the physical size and particle distribution of Au NPs can be easily tuned by sputtering technique. When the sputtering current increased from 30 mA (LPP1, Figure 6d) to 35 mA (HPP1, Figure 6e) with deposition period fixed to 1 s, we obtained the controllable Au NPs with lateral size increasing from ~3 to ~5 nm and vertical height increasing from ~5 to ~8 nm, respectively. For another, if we fixed the applied current to 30 mA and prolonged the deposit period to 2 s (LPP2, Figure 6f), the Au NPs maintain almost same physical size as LPP1 but the gap of adjacent deposited Au NPs sharply drops compared with LPP1.

3.2 The structure of the MoS$_2$-based photodetector

In order to investigate chemical composition of the prepared materials, the X-ray photoelectron spectroscopy (XPS) was employed. As shown in Figure 7, The peaks at 229.2 and 232.3 eV correspond to the doublet of Mo 3d$_{5/2}$ and Mo 3d$_{3/2}$, respectively. And the peaks of 226.3, 162.1 and 163.2 eV of the binding energy attach to the S 2s, S 2p$_{3/2}$ and S 2p$_{1/2}$, respectively [26, 27]. For Au 4f, the peak positions of 83.6 and 87.2 eV bind to the Au 4f$_{7/2}$ and Au 4f$_{5/2}$, indicating the Au NPs are directly introduced into the exfoliated MoS$_2$ sheet [28]. More importantly, the banding energies of Mo and S in Au decorated MoS$_2$ maintain the same values as that of bare MoS$_2$ sheet, indicating the introduction of Au NPs has non-influence on the crystal structures of exfoliated MoS$_2$ sheet.

Further, we used Raman spectroscopy of 532 nm laser to confirm the structural properties of the fabricated devices. For MoS$_2$, the difference of $E_{1g}$ and $A_{1g}$, $\Delta$, corresponding to in-plane and out of plane energy vibrations, is used to index the layer number of obtained MoS$_2$. Figure 8a shows the $\Delta$ of bulk MoS$_2$ and our exfoliated MoS$_2$ are 27.8 and 25.3 cm$^{-1}$, respectively, indicating that the thickness of bare MoS$_2$ is about 10 layers [29], which is highly consistent with the AFM results. After decorating Au NPs with MoS$_2$, it exhibits the $\Delta$ maintains nearly same as bare MoS$_2$ but the intensities obviously increase, shown in Figure 8b, c.

3.3 The performance of the MoS$_2$-based photodetector

The photoelectric performance of the fabricated photodetector was studied at room temperature, which applied a 980 nm laser source with controllable incident power. In order to produce the laser beam pulses, we combined an oscilloscope to...
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the incident laser source. Figure 9a shows the photocurrent plots of photodetector, where the illumination power and bias voltage are 1.60 mW and 2 V, respectively. Obviously, the Au NPs/MoS$_2$ heterostructure-based photodetector exhibits an ultra-high photocurrent (8.6 nA) compared with that of bare MoS$_2$-based photodetector (0.59 nA).

Figure 9b shows the plots of photocurrent vs. applied bias voltage ranging from 0.1 to 15 V. We obtained a photocurrent up to ~480 nA, when the applied incident laser power and bias voltage were 7.50 mW and 15 V, yielding an improved responsivity of 64 mA/W. Moreover, the I-V plots indicate the photocurrent owns a good linear relationship with applied bias voltage, when the illumination intensities tuned from 0.85 to 7.5 mW. For another, Figure 9c shows the dependence plots of photocurrent ($I_{ph}$, nA) on laser power irradiation ($P_{in}$, $\mu$W). It is found that the
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DOI: http://dx.doi.org/10.5772/intechopen.89251

...photocurrent follows a nonlinear dependence to the incident power intensity, $aP^b$, where $a$ and $b$ are constant for different bias voltage. For example, when the bias voltage are 15 V, the fitting $a$ and $b$ are $3.37 \times 10^{-2}$ and 1.34, respectively.

With respect to the stability of the photodetector, we performed the extended duration photocurrent measurements by periodically switching the incident laser under illumination of 3.20 mW at bias of 5 V, and the periods of both on and off state are 5 s. Figure 9d shows the photocurrents over 500 circles of continuous operation, exhibiting a well stability. Moreover, in order to characterize the response time for detecting infrared wavelengths of our designed device, we applied an oscilloscope in the process of laser excitation to produce laser pulses with a duration of 10 ms. Figure 9e shows the time-response of Au decorated MoS$_2$-based photodetector under illumination of 7.50 mW at bias of 15 V. It indicates that the rise time ($t_{\text{rise}}$) and fall time ($t_{\text{fall}}$) are 2.4 and 2.6 ms, respectively, which are significantly superb to that of other reported MoS$_2$ photodetectors.
3.4 The operational mechanism

In order to study the potential mechanism of our fabricated photodetector, we simulated the electric field distribution of Au decorated MoS$_2$ using finite element method in the infrared region. We assumed that the Au NPs with a gap of 6 nm and a diameter of 5 nm under illumination of a linearly polarized plane wave with an electric field amplitude of 1 Vm$^{-1}$ for the simulated model. And the thicknesses of MoS$_2$ sheet and incident laser wavelength were 6 and 980 nm, respectively. Figure 10a, b shows the cross-section of the simulated electric field distribution of Au NPs decorated MoS$_2$. Benefiting from the LSPR effect excited by the Au NPs, when the diameter matched to the incident wavelengths, the intensities of electric field at interfaces of air/Au/MoS$_2$, up to $\sim 3.96 \times 10^5$ V/m, are obviously higher than other districts. In comparison, interfaces of air/MoS$_2$ show a poor intensity of electric field, only about $6.72 \times 10^4$ V/m. This tendency can be also observed in the recent researches of Au NPs guided MoS$_2$ sheets for photo-detection [30]. We further experimentally proved the absorption by using UV-visible-NIR spectroscopy analysis. Figure 10c shows the absorption spectrum of bare MoS$_2$ sheet Au NPs decorated MoS$_2$. The normalized absorptance plots indicate that the Au NPs/MoS$_2$ is obviously enhanced than that of bare MoS$_2$ ranging from 700 to 1600 nm.

The above results and discussion clearly unveil the introduction of Au NPs plays a key role in enhancing the light matter interactions of MoS$_2$ with infrared wavelengths. The significantly improved sensitivity of the fabricated photodetector could be attributed to the LSPR effect, shown in Figure 10d, induced by the periodically aligned Au NPs, resulting in obvious improvement of local electric field. When the incident infrared wavelengths highly confined by the deposited Au NPs, the local electric field at the interface of Au/MoS$_2$ is greatly improved by the surface plasmon waves. Firstly, the Au surface plasmons effectively excite a
coupling effect at the interface of Au/MoS$_2$ for absorbing photons, yielding a much more photo-induced carriers to improve the photo sensing [30, 31]. Moreover, the additional local electric field generated by the LSPR effect of Au NPs accelerates the photogenerated carriers to separate for producing photocurrent [32, 33]. This facile method, tuning Au NPs by sputtering method to excite LSPR effect for fabricating the unique device structure, is expected to be practical applications in other 2D materials such as WS$_2$ and MoSe$_2$ [34–36], thus offers a new route on a variety of high-performance optoelectronic devices.

4. Conclusion

High performance photodetectors are very important in a lot of applications. We have successfully developed two simple methods to prepare plasmon-enhanced photodetectors. (i) Au nanoparticles (Au NPs) solution were directly spun coated onto the WS$_2$-based photodetectors. The performance has been enhanced by the LSPR of Au NPs, and reached an excellent high responsivity of 1050 A/W at the wavelength of 590 nm. (ii) Au NPs were deposited on MoS$_2$ by magnetron sputtering. The spectral response of pure MoS$_2$ was located in visible light and which was extended to near-infrared region (700–1600 nm) by Au NPs. Further, the responsivity reaching up to 64 mA/W when the incident light is 980 nm. These photodetectors achieved excellent responsivity and response speed. The results not only promote the development of high-performance photodetectors, but also provide a simplified method for the fabrication of other hybrid structure devices.

Acknowledgements

The project was supported by grants from the National Basic Research Program of China (No. 2015CB351905), the National Key Research and Development Program of China (No. 2016YFA 0302300, No. 2016YFA0200400), the National Natural Science Foundation of China (No. 61306105).

Conflict of interest

The authors declare no conflict of interest.
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Simple Preparations for Plasmon-Enhanced Photodetectors
DOI: http://dx.doi.org/10.5772/intechopen.89251

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