The role of plasmonic metal-oxides core-shell nanoparticles on the optical absorption of Perovskite solar cells

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Abstract
Among all the different methods to enhance the optical absorption of photovoltaic devices, the plasmonic effect is one of the most prominent and effective ways to capture more incident light and also provide good carrier dynamic management. Here, we systematically introduce spherical gold nanoparticles (Au NPs) with different radii in the absorber layer of perovskite solar cells (PSCs). The overall enhanced optical absorption of around 14.20% and 20.02% is achieved for incorporated monolayer and bilayer Au NPs, respectively, in the active layer compared to the pure perovskite layer. Moreover, we employ the metal (Au)-dielectric (TiO₂ and SiO₂) nanoparticles in the absorber layer. The optical absorption increases as the core-shell size decreases. The optical absorption elevates in both Au@TiO₂ core-shell and Au@SiO₂ core-shell 17.5% and 3.5%, respectively. These results support superior separation and transfer of charge in the existence of plasmonic NPs. In addition, this study presents a very sophisticated approach to the optical enhancement of PSCs and thus helps to boost the overall photovoltaic device performance.

Keywords Gold nanoparticle · Perovskite solar cell · Surface plasmons · Optical absorption · Core-shell nanoparticle
1 Introduction

The photovoltaic effect is the conversion of incident light into useful electricity. Among all solar cell materials, perovskite solar cell (PSC) is considered one of the most prominent and promised candidates in solar cells industries owing to their excellent properties such as cheaper cost, low-temperature chemical processing and fabrication (Alden Mostaan and Saghaei 2021; Ecija et al. 2012; Elumalai et al. 2016; Naghizade and Saghaei 2020), strong absorption of sunlight (Huang et al. 2017), higher mobility of carriers and low rate of non-radiative carrier recombination (Green et al. 2014). In addition, flexible PSCs of different colors can be fabricated effectively and are advantageous to utilize large and wide wavelength ranges. The important parts of PSCs are transparent conductive oxides (FTO, ITO), electron transport layer (ETL), absorber layer (perovskite), hole transport layer (HTL), and the metal contact (Kandjani et al. 2015). Miyasaka’s group presented the first-ever report and replaced dye-sensitized solar cells with perovskite (CH$_3$NH$_3$PbI$_3$) used as liquid sensitizer and achieved a power conversion efficiency (PCE) of 3.8% (Kojima et al. 2009). Kim et al. presented a successful report using perovskite as an active layer, and they also introduced Spiro-MeoTAD as a hole transport layer (HTL) and mesoporous TiO$_2$ as an electron transport layer (ETL). The PCE was achieved at 9.7%, almost three times higher than the one presented in the previous report (Kim et al. 2012). Moreover, PCE of around 22.1% has been investigated for PSCs and is highly expected to jump over 30% (Yang et al. 1979). The optimization of different parameters used in PSCs has become practicable by the rapid efficiency improvements. Each component has its role in improving optical absorption, PCEs, and stability. For instance, the perovskite layer thickness is a dominant factor that extensively affects the absorption of light and the charge separation in HTL and ETL, respectively (Liu et al. 2014). The thin layer of perovskite has not the capability to utilize more optical beams, but on the other hand, it provides a good pathway for charge separation. To resolve this hindrance an alternative solution is needed to enhance optical absorption and improve PSCs device performance without increasing the thickness of the perovskite absorber layer. In such circumstances, the surface plasmon resonance effect is one of the best solutions that utilize metal at the nanoscale embedded in PSCs; when the incoming light from the sun strikes the metal nanoparticle inside the absorber layer; as a result, electrons start shining on the surface of metals, such phenomenon of collectively shining of electrons on the surface of the metal is called localized surface plasmon resonance (LSPR) and thus enable the solar cells to harvest more light. These LSPRs also provide a strong electromagnetic field that leads to enhanced scattering cross-sections and extinction cross-sections for larger atoms (Atwater and Polman 2011). The effect of LSPR has extensively been studied in many photovoltaic devices besides PSCs, such as silicon-based solar cells (Derkacs et al. 2006; Tabrizi and Pahlavan 2020), dye-sensitized solar cells (DSSC) (Brown et al. 2011), and organic solar cells (OSC) (Vangelidis et al. 2018). Zhang et al. first investigated the plasmonic effect in PSCs using metal nanoparticles (NPs) and enhanced PCE to 9.5% as compared to controlled devices (8.4%) (Zhang et al. 2013). The plasmonic effect has also been investigated in thin organic solar cells. For example, Mola et al. introduced nickel sulfide (NiS) NPs in thin film organic solar cells absorber (Mola et al. 2021). The final solar cells device showed improved optical and electrical performance as compared to the controlled device by only 1% most favorable concentration of NiS NPs by weight. The proposed device showed higher absorption in NIR spectrum and also increased PCE. Similarly,
Thaver et al. introduced another very outstanding synthesis strategy by employing silver doped nickel oxides (Ag@NiO) nanocomposite in thin film organic solar cells (Thaver et al. 2021). The nanocomposite incorporated champion device showed better optoelectronics performance as compared to the pristine device.

Furasova et al. studied the effect of silicon NPs in PSCs that optimize the PCE up to 18.8% compared to the controlled device without NPs at 17.7% (Furasova et al. 2018). Batmunkh et al. reported the influence of gold nanostars on mesoporous TiO2 photoanode in PSCs; they observed the PCE increased from 15.19 to 17.72%. Moreover, enhanced optical absorption and reduced charged recombination were also investigated (Batmunkh et al. 2017). Aeineh et al. introduced multifunctional Au@SiO2 core-shell in PSCs and thus improved the device performance because of the plasmonic effect (Aeineh et al. 2017). The Au@SiO2 in PSCs is also capable of improved device stability. The stability is due to the SiO2 coating that acts as a shield and protects Au and the perovskite layer. However, a big gap still exists in wisely utilizing the plasmonic effect of nanoparticles with various geometries in PSCs structure. These plasmonic nanoparticles can dramatically change the performance of PSCs in terms of absorption, scattering, efficiency, and stability. Moreover, LSPR caused by these nanoparticles depends on the shape, size, and dielectric function procured by the embedded nanoparticles (Khan et al. 2019; Sui et al. 2019; Tabrizi et al. 2021).

In this paper, we performed the optical simulation of embedded Au NPs with different radii (g), Au@TiO2, and Au@SiO2 with different shell thicknesses (S_tk) in the absorber layer of PSC in the wavelength range, i.e., 300–800 nm. The plasmonic effect and the corresponding enhancement in the UV-Vis spectrum were systemically observed for each scheme by tailoring the radius (g) of Au NPs and shell thickness (S_tk). Then we further calculated the bandgap energy for each geometry (Au NPs, Au@TiO2, and Au@SiO2 modified inside the perovskite layer with the help of Tauc’s curve. In addition, we finally compared all the results and investigated that the overall optical enhancement in PSC is dedicated to Au@TiO2 as compared to simple Au NPs and Au@SiO2, thus providing a useful track for enhancement and improved stability of PSCs.

2 Physical structure

We first systemically studied the influence of Au NPs, Au@TiO2 and Au@SiO2 on the optical properties of PSCs; we used the finite element method (FEM) in all numerical simulations. Our proposed PSC model is depicted in Fig. 1a.

PSC structure normally consists of different functional layers like ITO/TiO2/Perovskite layer/Spiro-OMeTAD/Au. The thickness chosen for all the mentioned parameters were 150 nm, 60 nm, 250 nm, 80, and 60 nm, respectively. The refractive index assumed for glass substrate is 1.5. The dielectric function of gold used in the present work is according to the Johnson and Christy model presented in (Johnson and Christy 1972). The complex refractive index of ITO, TiO2, Spiro-OMeTAD, and perovskite are chosen from the literature for simulation, respectively (Filipič et al. 2015; Löper et al. 2015). In this simulating model, the entire simulation was performed in a unit cell. To obtain more precise and promised results, we employed tetrahedral meshing for the frequency-domain solver. The structure, as mentioned earlier, is composed of stacked layers obtained from distinct materials, which constitute a sandwich-type configuration. In this work, spherical-shaped plasmonic Au NPs...
I. Ullah et al. with different sizes were incorporated in TiO$_2$ and SiO$_2$ of various shell thicknesses (S$_{tk}$) of plasmonic nanoparticles is studied for the perovskite active layer shown in Fig. 1. An array of equally distributed NPs inside the absorber layer of PSCs is studied. Therefore, symmetry boundary conditions were practiced in the x- and y- axes. The regular two-dimensional patterned structure is shown in Fig. 2 (a, b). To make our model more practical, the background environment is filled with air. The z-axis is kept “open (add space)” for an incoming electromagnetic wave to enable the perovskite layer to absorb the incident light. The simulated model shows that the incident electromagnetic wave propagates in the z-direction while x- and y- directions are dedicated to magnetic and electric fields, respectively.

Radiofrequency (RF) module is used by enabling the scattered field formulation to model the optical properties of the full stack proposed device. The standard solar spectrum AM1.5 and irradiance 100 W/m$^2$ is employed in the photovoltaic device. Moreover, the following mathematical equation is used to calculate optical absorption coefficient A(λ):

\[ A(\lambda) = \frac{\text{absorption}}{\text{area}} \]

\[ A(\lambda) = \frac{\sum_{i=1}^{n} \text{absorption}_i}{\text{area}} \]

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where lambda (\(\lambda\)), R, and T represent the wavelength, reflection, and transmission of light, respectively (Tavakoli et al. 2019). The interaction of electromagnetic wave (EMW) with metal NPs that tend to excite electrons results in further generation of surface plasmon polariton. Maxwell’s equations are used to calculate it mathematically when EMW propagates between metal and dielectric.

3 Results and discussions

Initially, we have studied the influence of Au NP, incorporated in the active layer of PSC, on optical absorption, as shown in Fig. 3a. The size of Au NP was first chosen as 30 nm and then increased to 40 and 50 nm, respectively. The results showed that as the Au NPs size increases, the absorption also increases compared to the neat PSC (without Au NPs), as shown in Fig. 3b. This optical absorption enhancement is mainly because of the strong near-field and far-field scattering caused by plasmonic NPs. The enhanced and broadband absorption, i.e., 550–800 nm investigate due to the dielectric properties of Au NP (Hajjiah et al. 2018). These novel simulations further verified the experimentally based results in term of optical absorption in the wave range from 650 to 800 nm and higher photon current density (Shalan et al. 2017). To further validate our results, we obtained the optical bandgap energy using the Tauc curve (Rehman et al. 2021; Roy and Botte 2018). The bandgap energy decreases from 1.69 eV (without Au NPs) to 1.61 eV, 1.53 eV and 1.51 eV as the nanoparticles size increased from 30 to 50 nm, respectively as shown in Fig. 3c.

As a result, the energy required to excite electrons is reduced, so electrons will easily jump from the valence band to the conduction band. Moreover, we further analyzed the optical absorption of the active layer with and without Au NPs in the simulations. Therefore, we calculated the percent amount of absorption enhancement of the absorber layer by using Au NPs with various radii, taking the absorption value for the active layer without plasmonic effect as a reference for comparison. The corresponding enhancement in absorption values (6.2%, 10.92%, and 18.32%) for the diameters 30 nm, 40 nm, and 50 nm, respectively is shown in Table 1.

In addition, we further explored the influence of bilayer Au NPs placed inside the absorber layer in the z-direction. The size of 50 nm (AuNPs) was chosen for the bilayer; this is the extreme size of NPs because there was not enough space to put more NPs in the unit space designed. The enhancement in UV-Vis spectrum for bilayer Au NPs incorporated in the perovskite layer is depicted in Fig. 4a. The optical absorption spectrum of the bilayer Au NPs embedded perovskite layer is compared with single Au NPs incorporated and without Au NP in the absorber layer. The results showed further enhancement in optical absorption up to 91.46%, as reported in Table 1. The percent increment is also investigated in the case of bilayer Au NPs up to 6% and 14% compared to single NPs embedded in the active layer and pure perovskite, respectively. The Tauc curve is depicted in Fig. 4b. The Tauc curve showed that the decrease in bandgap energy strongly supports our previous results because of the optimization of Au NPs and LSPR effects.
4 Effect of metal-dielectric core-shell thickness on optical absorption of PSC

4.1 Effect of Au@TiO$_2$ shell thickness

In this section, we have studied the influence of Au@TiO$_2$ shell thickness ($S_{th}$) on the optical properties of PSCs. The schematic of Au@TiO$_2$ embedded in the perovskite layer is shown.
in Fig. 5a. The optical absorption spectrum, as shown in Fig. 5b suggests that the $S_{th}$ has a positive effect on the optical performance of PSCs. The broad-spectrum is achieved between 630 and 800 nm as the $S_{th}$ decreases; at the same time, the shell layer improves the stability of NPs and minimizes the recombination effect by providing safety to avoid unnecessary interaction between NP and absorber layer (Fan et al. 2017). Moreover, the converted hot electrons originated from the absorbed photons are linked to the Landau damping, which happens when the phase velocity of electromagnetic approaches that of the plasma particles and possesses superior coupling with each other (Lee et al. 2014). When these hot electrons arrive at the perovskite layer, they provide a very sophisticated path to utilize more useful energy around the electromagnetic waves and thus improve the carrier dynamics in the device. These results further confirmed and matched with the experimental results, the experimental based also showed better optical and electrical performance by incorporating Au@TiO$_2$ core shell (Luo et al. 2017). In addition, the Tauc curve, as shown in Fig. 5c, also provided information about bandgap and the comparison of different shell $S_{th}$ used in the simulation, the bandgap energy of PSC without Au@TiO$_2$ is about 1.89 eV, with embedded only TiO$_2$ inside the absorber layer is 1.85 eV and with Au@TiO$_2$.

### 4.2 Effect of Au@SiO$_2$ shell thickness

Here we performed the detailed simulations of Au@SiO$_2$ incorporated in the perovskite layer as depicted in Fig. 6a, and their impact on the optical absorption spectrum of PSC. The absorption spectrum for this scheme is shown in Fig. 6a. A very slight increase is observed in the optical spectrum of PSCs with Au@SiO$_2$, which means that the silica shell has no prominent impact on the optical enhancement, or the shell layer has a very small influence on the optical properties due to the dielectric properties of surrounding NP after coating. However, dielectric silica offered both thermal and structural stability. The enhancement, in this case, is mainly due to the increased Au NPs size (Zhang et al. 2013). These findings better supports the fabrication work done and experimental based results also superior optical absorption by embedding Au@SiO$_2$ in the perovskite layer (Chandrasekhar et al. 2018). The Tauc’s curve in Fig. 6b further validated the previous result. There is no such difference obtained in the bandgap energy by using the Au@SiO$_2$ metal-dielectric core-shell. Moreover, the enhancement in the absorption with and without assuming Au@SiO$_2$ is shown in
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Table 3. The maximum enhancement of 3.5% was achieved in the overall optical absorption for Au@SiO$_2$ for $S_{tk}$ of 20 nm. While in the other two cases for $S_{tk}$ of 30 and 40 nm, a very small enhancement was observed at 1.04% and 2.6%, respectively.

5 Comparison of absorption enhancement of au NPs, Au@TiO$_2$, and Au@SiO$_2$ core-shell

Finally, we compared all the simulated schemes embedded in the perovskite layer i.e., Au NPs, Au@TiO$_2$, and Au@SiO$_2$ core shells. In the first scenario, we introduced monolayer and bilayer Au NPs embedded in the perovskite layer and investigated about 14.02% and 20.20% optical enhancement due to the LSPR effect, near field enhancement, and far-field scattering. In the second case, we introduced plasmonic Au NPs in pure TiO$_2$ to form core-shell nanostructure; the results are shown in Fig. 7a. a redshift, as well as enhancement in optical absorption, was observed by wisely incorporated Au NPs in TiO$_2$ nanoparticle to enhance the index of refraction of the insulating medium around the Au NPs. Because pure
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TiO\textsubscript{2} has the capability to absorb less than 5% of the solar spectrum (Li et al. 2018). We found a superior optical enhancement of around 17.23% for embedding Au@TiO\textsubscript{2} core-shell in the perovskite layer as shown in Fig. 7a. Finally, in the case of incorporating Au@SiO\textsubscript{2} core-shell in the perovskite layer, a very slight redshift and very small 3.5% optical enhancement were observed in the absorption spectrum of PSC for the same S\textsubscript{tk} of 20 nm in both cases. The small increment in optical absorption is due to the low dielectric constant of silica as compared to the titania dielectric medium. Moreover, the reduced bandgap calculation from Tauc’s curve also confirmed our previous outcomes. Similarly, the same concept was followed for the bandgap calculation of Au@TiO\textsubscript{2} core-shell and Au@SiO\textsubscript{2} core-shell. The decrease in bandgap achieved as the shell thickness decreases, for minimum shell thickness

| Structure                  | Values of normalized absorption of neat PSC (%) | Values of normalized absorption of PSC with core-shell (%) | Enhancement in normalized absorption (%) |
|---------------------------|-----------------------------------------------|-----------------------------------------------------------|------------------------------------------|
| Pure Perovskite           | 76.84                                         | -                                                         | 0                                        |
| Pure TiO\textsubscript{2} | 76.84                                         | -                                                         | 0                                        |
| Au@TiO\textsubscript{2} (40 nm) | -                        | 84.26                                                     | 9.52                                     |
| Au@TiO\textsubscript{2} (30 nm) | -                        | 88.84                                                     | 15.64                                    |
| Au@TiO\textsubscript{2} (20 nm) | -                        | 90.02                                                     | 17.23                                    |

The bandgap is reduced to 1.6 eV, 1.55 eV, and 1.52 eV for shell thickness S\textsubscript{tk}=40 nm, 30 nm, and 20 nm, respectively. Moreover, the percentage increase in the UV-Vis spectrum is listed in the Table 2. The normalized absorption of neat PSC is the same as the embedded only spherical TiO\textsubscript{2} NPs while using Au@TiO\textsubscript{2} inside the absorber layer; the overall absorbance increased to 9.52%, 15.64%, and 17.23% for different S\textsubscript{tk} 40 nm, 30 nm, and 20 nm respectively. The enhancement of the UV-Vis spectrum in PSC is because of the transfer of electrons from Au NPs to TiO\textsubscript{2} and is also dedicated to the light scattering phenomenon in metallic NP (Cheng et al. 2017; Yuan et al. 2015).

Fig. 6 Monolayer Au@SiO\textsubscript{2} core-shell with different sizes, i.e., 40 nm, 30 nm and 20 nm (a) UV-VIS spectrum, (b) Tauc’s curve for bandgap calculations
In conclusion, we theoretically designed plasmonic and metal-dielectric core-shell nanostructure put in the active layer of PSCs and very wisely examined the plasmonics effect. Our optimized approach showed that the plasmonic effect strongly relies on the NPs size, shape, geometry, and the metal NPs and dielectric interaction. Firstly we have reported the enhancement in optical absorption of PSCs by inserting the Au NPs with different radii in the absorber layer due to the plasmonic effect. The Au NPs with increasing radii showed about 14% (g = 50 nm) improved optical enhancement and achieved a broad spectrum in PSCs as compared to neat PSCs without plasmonic effect. Secondly, we have also investigated the effect of $S_{tk}$ of Au@TiO$_2$ and Au@SiO$_2$ incorporated in the active perovskite layers and their influence on the optical absorption of PSCs. Our findings depicted that the $S_{tk}$ has a positive impact on the UV-Vis absorption of PSCs. The UV-Vis absorption increased as the size of the shell decreased. Finally, we compared the UV-Vis absorption of Au@TiO$_2$ (S$_{tk}$ = 20 nm) of both Au@TiO$_2$ and Au@SiO$_2$ core-shell, the optimized reduced bandgap 1.52 eV investigated as compared to pure perovskite.

### 6 Conclusions

In conclusion, we theoretically designed plasmonic and metal-dielectric core-shell nanostructure put in the active layer of PSCs and very wisely examined the plasmonics effect. Our optimized approach showed that the plasmonic effect strongly relies on the NPs size, shape, geometry, and the metal NPs and dielectric interaction. Firstly we have reported the enhancement in optical absorption of PSCs by inserting the Au NPs with different radii in the absorber layer due to the plasmonic effect. The Au NPs with increasing radii showed about 14% ($g = 50$ nm) improved optical enhancement and achieved a broad spectrum in PSCs as compared to neat PSCs without plasmonic effect. Secondly, we have also investigated the effect of $S_{tk}$ of Au@TiO$_2$ and Au@SiO$_2$ incorporated in the active perovskite layers and their influence on the optical absorption of PSCs. Our findings depicted that the $S_{tk}$ has a positive impact on the UV-Vis absorption of PSCs. The UV-Vis absorption increased as the size of the shell decreased. Finally, we compared the UV-Vis absorption of Au@TiO$_2$.
and Au@SiO₂ for the same radii. The best optimum optical absorption is achieved in the case of Au@TiO₂ rather than in Au@SiO₂. The smart tailoring optical properties of Au NPs and their coupling with core-shell nanostructure open huge opportunities for future aspects to improve the technology in terms of chemical and thermal stability.

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**Declarations**

**Conflict of interest** There are no conflicts to declare.

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