

# Strong Absorption in a 2D Materials-based Spiral Nanocavity

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**Abstract**— Recent investigations of semiconducting two-dimensional (2D) transition metal dichalcogenides have provided evidence for strong light absorption relative to its thickness attributed to high density of states. Stacking a combination of metallic, insulating, and semiconducting 2D materials enables functional devices with atomic thicknesses. While photovoltaic cells based on 2D materials have been demonstrated, the reported absorption is still just a few percent of the incident light due to their sub-wavelength thickness leading to low cell efficiencies. Here we show that taking advantage of the mechanical flexibility of 2D materials by rolling a molybdenum disulfide ( $\text{MoS}_2$ )/graphene (Gr)/hexagonal Boron Nitride (hBN) stack to a spiral solar cell allows for solar absorption up to 90%. The optical absorption of a  $1\text{ }\mu\text{m}$  long hetero-material spiral cell consisting of  $\text{MoS}_2$ , graphene and hBN is about 50% stronger compared to a planar  $\text{MoS}_2$  cell of the same thickness; although the ration of the absorbing material, here Gr and  $\text{MoS}_2$ , relative to the cell volume is only 6%. We anticipate these results to provide guidance for photonic structures that take advantage of the unique properties of 2D materials in solar energy conversion applications.

## 1. INTRODUCTION

The most widely installed material for solar cells are amorphous, poly or single crystalline Silicon, and more recently II-VI semiconductors such as cadmium telluride have been widely studied and utilized in the photovoltaic industry. The recent isolation of two-dimensional (2D) materials [4, 5], and their combination in vertical [1, 6, 9, 24] and horizontal functional systems [7, 8] has provided opportunities to form heterostructures that are attractive candidates for solar energy conversion applications. For instance, group VI transition metal dichalcogenides (TMD) are 2D crystals that can exhibit semiconducting behavior. Such materials are constructed by the formula  $\text{MX}_2$  ( $M$  = metal, e.g., Mo or W;  $X$  = semiconductor, e.g., S, Se, or Te) and are structured such that each layer consists of three atomic planes: a lattice of transition metal atoms sandwiched between two lattice of chalcogenides. There is strong covalent bonding between the atoms within each layer and adjacent layers are held together by weak van der Waals forces. These atomically thin heterostructures of semiconducting TMDs allow for surprisingly strong light-matter interactions, which can be utilized for harvesting sunlight via absorption and photovoltaic effects [1]. Previously TMD/Gr bilayers of vertical heterostructure stacks were studied to improve photocurrent extraction [1, 9, 24] in photovoltaic and photodetector applications. These heterostructures can utilize effective combination of good solar spectrum absorption of some TMDs such as  $\text{MoS}_2$  and superior mobility of graphene (i.e.  $200,000\text{ cm}^2/\text{Vs}$  for suspended graphene [18]) in a Schottky barrier solar cells. Although a few layers of both graphene and  $\text{MoS}_2$  are visually transparent, they are promising sunlight absorbers due to their large absorption per thickness and high density of states. Classically the amount of light absorbed in flat photovoltaic cells is proportional to the film thickness. However, creating an increased optical path length significantly reduces the amount of required photoactive materials. This can be achieved via light management architectures such as planar metamaterial light-director structures, Mie scattering surface nanostructures, metal-dielectric-metal waveguide or semiconductor-dielectric-semiconductor slot waveguides [21]. Here we investigate a three-dimensional solar cell structure based on a variety of stacked 2D materials each with a functional purpose (Figs. 1(a) and 1(b)); by rolling a stack of graphene, semiconducting  $\text{MoS}_2$ , and an electrically insulating 2D material this structure creates an absorbing cylinder forming a lightconcentrating optical cavity as we discuss below. This structure is motivated by deploying the mechanical flexibility of 2D materials to enable a multilayer solar cell without the necessity to contact each of the layers separately. The electronic barrier layer needs to be flexible, wide bandgap, and optically transparent insulator to prevent electron and holes generated in a  $\text{MoS}_2$ /Gr heterojunction from recombining in their adjacent stacked layer. Hexagonal boron nitride (hBN) is employed as barrier layer since it is an isomorph of graphene, optically transparent, electrically an insulator with a wide bandgap of about 5.9 eV, and it reduces the traps of  $\text{MoS}_2$  layers indicated by improved mobilities [6]. Furthermore, a trilayer  $\text{MoS}_2$  with a bandgap of 1.6 eV [11] was chosen over a monolayer since bulk behavior

is not expected to emerge beyond stacking of 3–4 monolayers [9,19], whereas mobility improves with the number of layers [6]. In this study we contrast two absorbers, namely, the rolled up Gr/TMD/hBN “spiral” cell (Fig. 1(a)), and a metal-cladded and metal inner-post “hetero-shell” cell (Fig. 1(b)). The reason behind studying both is that as to separate the effect of the material (i.e., TMD) absorption from any optical nano-cavity effects the hetero-shell device might exhibit. For the latter design, the stack rolls around a core metallic rod and is then coated by another metallic shell, where both metals are the electrical cell’s contacts. The core metallic rod with a low work function (aluminum) is in contact with the graphene, whereas the shell contact with a higher work function (gold) contacts to the MoS<sub>2</sub> layer towards establishing selective cell contacts. In our experiment, we numerically investigate light absorption deploying finite difference time-domain (FDTD) techniques. Comparing this spiral design to previously reported TMD based photovoltaic cells and thick (1  $\mu\text{m}$ ) planar MoS<sub>2</sub> solar cells, we obtain a relative absorption enhancement which serves as a reference.

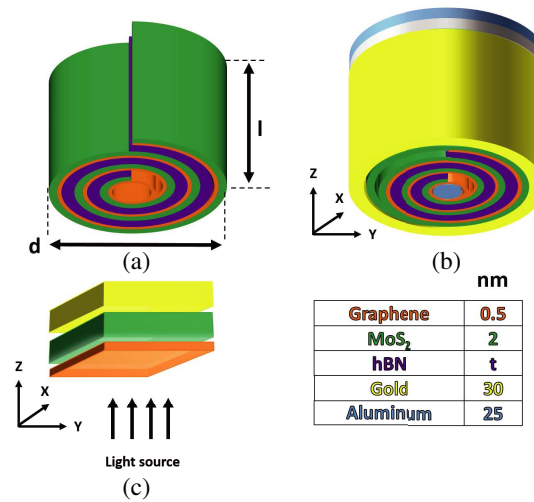


Figure 1: (a) Schematic of “spiral cell structure”; parameters,  $d$ ,  $l$ , and  $t$  stand for diameter of the roll, length of the cylindrical structure, and thickness of the hBN layer. (b) “Core-shell structure” of the spiral solar cell. Back reflector is connected to the core contact. Gold (aluminum) is chosen as a shell (core) selective contact. (c) Planar MoS<sub>2</sub>/Gr solar cell converting vertically incident photons into electron-hole pairs.

To obtain the absorption efficiency and spectral current density of the spiral cell, we perform 3D FDTD simulations to solve Maxwell’s equations as a function of time and then perform a Fourier transformation. For efficiency, this time-domain method covers a wide frequency range in a single simulation run. A multi-coefficient model was used to represent the complex refractive index of a trilayered MoS<sub>2</sub> [11], the Graphene monolayer [12], and hBN [13]. The approach is to send a broadband, normally incident plane wave pulse (300 to 800 nm) on the spiral structure or the planar solar cell (Fig. 1). The spiral cell was tested under both transverse electric (TE) and transverse magnetic (TM) polarization. The length of spiral structure studied are between a convenient small size for heavily meshed simulations and the length in which we obtain the highest absorption (0.5  $\mu\text{m}$  and 3  $\mu\text{m}$ ); and their diameter range from 100 nm to 2  $\mu\text{m}$  as the effect of hBN thickness on spiral cell is examined. We analyze an isolated single spiral cell and planar cell for which a perfectly matched layer and periodic boundary condition were selected, respectively, in the  $x$  and  $y$  direction. Note, a perfectly matched layer is assumed in propagation direction to absorb any back reflected waves. Furthermore, a power monitor surrounds the entire cell to obtain the net flow out of the simulation domain ( $P_{out}$ ). The light source is placed inside the power analysis volume, and the absorption ( $A$ ) is obtained by  $A = 1 - P_{out}$ . The absorption per unit volume can be calculated from the divergence of the pointing vector:

$$P_{abs} = -0.5R(\vec{\nabla} \cdot \vec{p}) = -0.5\omega E(\omega)^2 \varepsilon''(\omega)$$

where  $\omega$  is the angular frequency,  $\varepsilon''(\omega)$  the imaginary part of permittivity function, and  $E(\omega)^2$  the electric field density. Hence, to calculate the spatial and frequency function of the absorption, we only need to know electric field intensity and imaginary part of the complex refractive index.

For a solar cell, of relevance is the current density ( $J$ ), which requires knowledge of the optical generation rate ( $G$ ), which is given by  $G(\omega) = \frac{P_{abs}}{\hbar\omega} = \frac{-E(\omega)^2 \varepsilon''(\omega)}{2\hbar}$  from which we obtain the spectral current density  $J_{SC}(\lambda) = \alpha(\lambda)IQE(\lambda)\Gamma_{AM1.5}\lambda/1.24$ , where  $\alpha$  is the absorption efficiency,  $\lambda$  is the wavelength, and  $\Gamma$  is the spectral irradiance of the AM 1.5G spectrum at one sun solar intensity (data taken from [14]). Note, the internal quantum efficiency (IQE) is assumed to be unity for calculating the short circuit current ( $J_{SC}$ ) is obtained by integrating  $J_{SC}(\lambda)$  over the wavelength range of 300–800 nm, namely

$$J_{SC} = \int \frac{e\lambda}{hc} EQE(\lambda) \Gamma_{AM1.5}(\lambda) d\lambda.$$

The spectral scan displays resonant-like fringes in the absorption spectrum (Fig. 2(b)). We also find a higher visibility for the hetero-shell cell with the metal cladding compared to the dielectric spiral design, which can be understood from an optical field confinement inside the structure. The resonances themselves point towards the spirals resembling a nanowire like structures being a lossy Fabry-Perot cavity [22, 23], which we confirm via (i) investigating the modal features of this cavity (Figs. 2(c) and 2(d)), and (ii) analyzing their frequency profile (Fig. 3(c)). Regarding (i), the  $x$ - $y$  mode profile (i.e.,  $x$  in Fig. 2(a)) indicates a dipole for larger wavelength, which turns into quadruples and double-quadruples for blue shifting the resonance wavelength (6 to 1 in Figs. 2(c) and 2(d)). In addition the cavities' standing waves can be seen in the cross-sectional modal profile (i.e.,  $x'$  in Fig. 2(a)), where the mode spacing decreases with wavelength as expected (Figs. 2(c) and 2(d)). The in Figure 2b observed higher Q-factor of the hetero-cell over the spiral cell is clearly visible in the crosssectional mode profiles as distinct power density lobes. The apparent focusing effect in the  $z$ -direction might be connected to a changing (i.e., increasing) local effective index as experienced by the wave traveling in positive  $z$ , however more details are needed to confirm this. Regarding (ii), analyzing the resonance peak spacing from Fig. 2(b) and relating them to the inverse of the cavity length allows to test the Fabry-Perot cavity hypothesis analyzing its spectral mode spacing (Fig. 3(c)). Finding the results along a straight line confirms that the hetero-cell is a

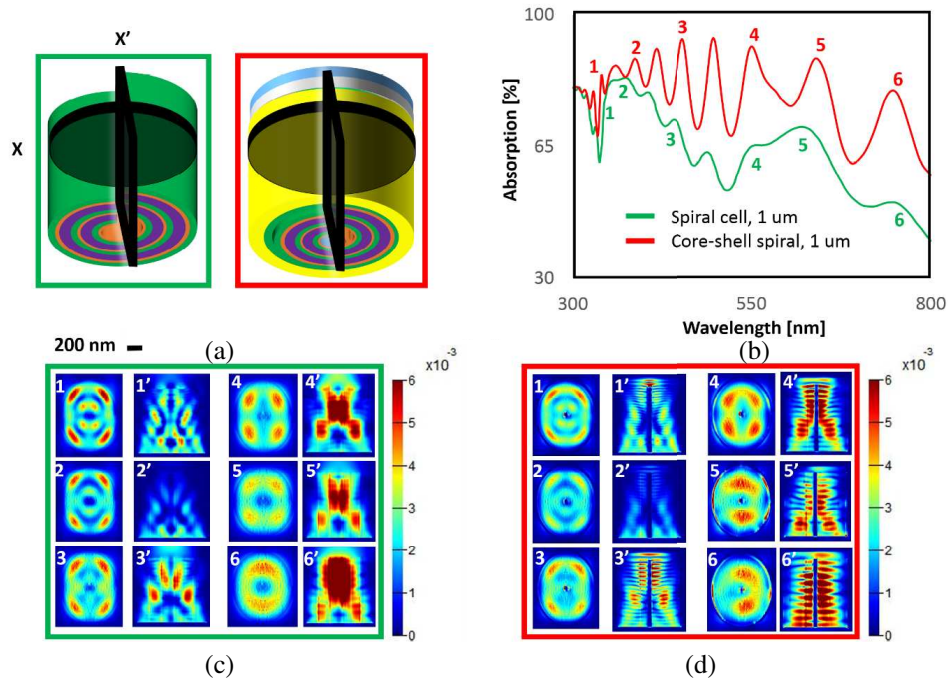


Figure 2: (a) Schematic of spiral cell (left) and core-shell structures (right). Horizontal and vertical cross sections labeled by  $X$  and  $X'$  are monitors that record the power profiles shown in (c) and (d). (b) Absorption efficiency of spiral cell and core-shell structure. (c) Power profile of absorption efficiency peaks from 1 to 6 for spiral cell structure correspond to 344, 375, 441, 550, 627, and 744 nm wavelength respectively. (d) These wavelengths correspond to wavelength of peaks of the core-shell structure at 340, 388, 455, 550, 640, and 750 nm.

nanoscale cavity. This discussion suggests that the spiral structure behaves like a circular dielectric waveguide where the end facets act as reflecting mirrors [15, 16]. Neglecting dispersion we use  $m = 2nL/\lambda$ , where  $m$  is the mode (an integer) and  $L$  is the length of the spiral cell (cavity),  $n$  is the effective refractive index, and  $\lambda$  is the wavelength. The spiral cell structure supports mode numbers between 5 and 13 that correspond to 9 visible peaks of absorption efficiency (Fig. 2(b)). This is supported by longitudinal mode profiles recorded by power monitors in Fig. 2, which demonstrates more interference visibility at core shell structure for all monitored wavelengths.

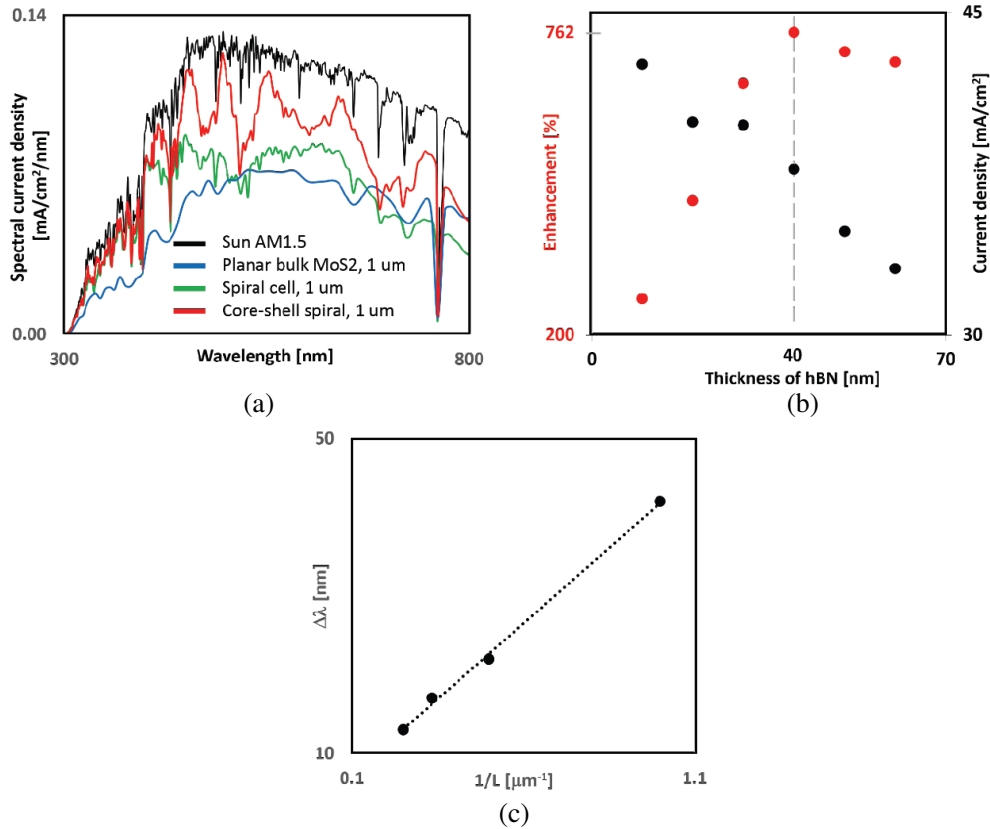


Figure 3: (a) Spectral current density of all three structures. Green, red, and blue curves correspond to Figure 1a,b,c respectively.  $d = 1 \mu\text{m}$ ,  $l = 0.8 \mu\text{m}$  (b) Optimization of hBN thickness to achieve maximum current density to photoactive material ratio. (c) Mode spacing versus inverse nanowire length.

The spectral current density of a  $1 \mu\text{m}$  thick planar MoS<sub>2</sub> cell, the spiral cell, and the core-shell structure are compared (Fig. 3(a)). We find the spectral current density to be higher for the spiral structures over almost the entire investigated wavelength range. This is mainly because spiral structures have lower surface reflection due to lower effective refractive index of spiral structures compared to planar MoS<sub>2</sub> structure. Core-shell structure has an even higher absorption efficiency than the spiral cell, because the back reflector at the end of core-shell structure allows for double pass of light. The overall current densities of the planar cell, spiral cell, and core-shell structure integrated over wavelengths of 300 to 800 nm are 25.5, 29.5, and 37.2 mA/cm<sup>2</sup> respectively. The current density of the spiral (core) cell shows 16 (46)% enhancement compared to the planar structure which is expected, because the spectral current density is higher for spiral structures over almost the entire wavelength.

Note, that although the spiral length ( $l$ ) was kept constant ( $1 \mu\text{m}$ ) this does not imply that the same amount of photoactive material was used; for example in a particular simulation, the thickness of monolayer graphene, trilayer MoS<sub>2</sub>, and few layer hBN are set to 0.5 nm, 2.0 nm and 35.0 nm, respectively; and for the planar cell thickness of MoS<sub>2</sub> is  $1 \mu\text{m}$ . Thus, the absorbing materials (graphene and MoS<sub>2</sub>) occupy only 6% of the total volume of spiral cells. This means that the ratio of solar energy absorption to volume of photoactive material was improved by  $\sim 767\%$  compared to a bulk MoS<sub>2</sub> photovoltaic cell of the same size. We name this ratio “enhancement” and define

it as

$$\frac{\alpha_{\text{spiral cell}} - \alpha_{\text{bulk cell}}}{\alpha_{\text{bulk cell}}} \div \frac{t_{\text{MoS}_2} + t_{\text{Gr}}}{t_{\text{MoS}_2} + t_{\text{Gr}} + t_{\text{hBN}}},$$

where  $\alpha$  denotes absorption and  $t$  refers to the respective physical layer thickness. This enhancement is proportional to the absorption efficiency of the cell and thickness of hBN layer. However, increasing the thickness of the hBN layer, the absorption efficiency decreases due to a reduction in the amount of absorbing material (Fig. 4). Hence, to optimize the ratio of absorption enhancement to relative volume of photoactive material, the number of core shell structures with different hBN thicknesses are analyzed (Fig. 3(b), Fig. 4). For core-shell structures with 5 rings and 500 nm in length, the optimized enhancement is 762% for an hBN of 40 nm thickness. The dielectric strength of ultrathin hBN was previously studied to be 7.94 MV/cm [17], which corresponds to a breakdown voltage of 9 volts for a 10 nm thin layer of hBN. This is sufficient to prevent generated excitons to channel from one junction to another.

## 2. CONCLUSION

We have investigated a novel photovoltaic absorber and successfully demonstrated that rolling 2D materials into 3D structures can significantly improve their photo absorption compared to atomically thin or even bulk configurations. We optimized spiral solar cell design for the largest absorption, and optimized the hBN thickness in particular to achieve maximum absorption to photoactive material ratio. We can estimate a lower limit and a reasonable limit for power conversion efficiency of one of the spiral core-shell structure using current density ( $J_{SC}$ ) of 37.2 mA/cm<sup>2</sup> for the cell with length of 1  $\mu$ m and radius of 400 nm. For the lower limit and reasonable limit, the open circuit voltage ( $V_{OC}$ ) of 0.1% and 0.5 V [9], and fill factor (FF) of 0.3 and 0.7, are assumed respectively. In both cases IQE of unity and input power ( $P_{in}$ ) of 475 W/m<sup>2</sup> (input power in our simulation range of 300 nm to 800 nm) are assumed. The efficiency  $\eta$  is then calculated from:

$$\eta = \frac{FF \times J_{SC} \times V_{OC}}{P_{in}}$$

Resulting in an overall power conversion efficiency range of 2 to 27 %. Low open circuit is one of the main drawbacks for this solar cell. One way to overcome this barrier is use of different combination of 2D materials such as aiming for type two hetero junction such as MoS<sub>2</sub>/WSe<sub>2</sub>. Furthermore, recent efforts on enabling horizontally tunable bandgap TMDs [7, 8] utilized in this structure could lead to a multi junction photovoltaic cell that only requires two contacts.

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