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Semiconductor Two-dimensional PdQ₂ (Q=S, Se) monolayer: Strain Modulating electronic band gaps and SQ Efficiencies

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Abstract

We studied the physical, electronic transport and optical properties of a unique pentagonal PdQ₂ (Q= S, Se) monolayers. The dynamic stability of 2D - wrinkle like - PdQ₂ is proven by positive phonon frequencies in the phonon dispersion curve. The optimized structural parameters of wrinkled pentagonal PdQ₂ are in good agreement with the available experimental results. The ultimate tensile strength (UTHS) was calculated and found that, penta-PdS₂ monolayer can withstand up to 16% (18%) strain along *x* (*y*) direction with 3.44 *GPa* (3.43 *GPa*). While, penta-PdSe₂ monolayer can withstand up to 17% (19%) strain along *x* (*y*) direction with 3.46 *GPa* (3.40 *GPa*). It is found that, the penta-PdQ₂ monolayers has the semiconducting behavior with indirect band gap of 0.94 and 1.26 *eV* for 2D-PdS₂ and 2D-PdSe₂, respectively. More interestingly, at room temperature, the hole mobility (electron mobility) obtained for 2D-PdS₂ and PdSe₂ are 67.43 (258.06) *cm*² *V*⁻¹ *s*⁻¹ and 1518.81 (442.49) *cm*² *V*⁻¹ *s*⁻¹, respectively. In addition, *I-V* characteristics of PdSe₂ monolayer show strong negative differential conductance (NDC) region near the 3.57 *V*. The Shockly-Queisser (SQ) efficiency parameters of PdQ₂ monolayers are also explored and the highest SQ efficiency obtained for PdS₂ is 33.93% at -5% strain and for PdSe₂ is 33.94% at -2% strain. The penta-PdQ₂ exhibits high optical absorption intensity in the *UV* region, up to 4.04×10^5 (for PdS₂) and 5.28×10^5 (for PdSe₂), which is suitable for applications in optoelectronic devices. Thus, the ultrathin PdQ₂ monolayers could be potential material for next-generation solar-cell applications and high performance nanodevices.

1. Introduction

Atomically thin two-dimensional (2D) structures like, honeycomb lattice graphene [1, 2], boron nitride (BN) [3, 4], transition metal dichalcogenides (TMDs), group - IV, V [5, 6] elements has received attention due to their potential applications in the field of electronics, optoelectronics, solar cell, energy harvesting, and biomedical technologies with the high possibilities of replacing traditional 2D materials [1-7]. Among the 2D materials, if targeting application in nanodevices, MX_2 ($\text{M} = \text{Mo}, \text{W}, \text{V}, \text{Ti}, \text{Pd}, \text{Pt}$; $\text{X} = \text{S}, \text{Se}, \text{Te}$) type TMDs are technologically favourable materials [7-9]. Nowadays, an endless research is going on traditional TMDs layered materials, which is in demand of high mobility, high stability in air and strain modulated bandgap. Of these, many of TMDs has been already synthesized via the various method of exfoliation [10-14]. Interestingly, palladium based dichalcogenides has brought 2D with buckled or puckered pentagonal structures under the spotlight because of its unique lattice arrangement and fascinating properties such as fine tuning their physical, electronic, thermal and optical properties.

Gronvold et al., studied the $\text{PdS}_2/\text{PtS}_2$ and concluded that noble metals (Pd and Pt) would be a layered structure with sulphur atoms [15, 16]. Afterwards, in 2015 Wang et al., theoretically reported that 2D PdS_2 is semiconductor with moderate indirect band gap up to $\sim 1.60 \text{ eV}$, which is very significant in fabrication of nanodevices [17]. Moreover, Cheng et al., [18] have developed ultrafast Yb doped fiber laser using few layers of PdS_2 and indicated that layered PdS_2 can be a favourable candidate for photonics application. Some work have also reported on the layered PdSe_2 via method of mechanical exfoliated, atomic-resolution scanning transmission electron microscopy (AR-STEM) and chemical vapor deposition (CVD) [14, 19-24]. For Example, Akinola et al., [24] had fabricated few layers puckered pentagonal PdSe_2 by AR-STEM method and demonstrated that, PdSe_2 exhibits good ambipolar semiconducting nature with room temperature electron-apparent field-effect mobility (up to $\sim 158 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$) and its stability is remain same up to 60 days. It has been noted that having high mobility (electron/hole) and band gap between $1.2 \sim 1.9 \text{ eV}$ of TMD, it makes them suitable for the FETs (field effect transistors) and CMOS applications, respectively [25-28].

Recently, Weiting and their research group [29] had successfully synthesized 2D PdSe_2 nanosheets on 300 nm SiO_2/Si substrate and claimed that it is really promising material to make infrared photodetector due to its high-photoresponsivity ($\sim 660 \text{ A.W}^{-1}$ under 914 nm

laser) in harsh condition, which may be benefitted in military field for night-time detection because of its ability to work at night as well as in bad weather conditions. Beside these, few other reports theoretical are also available on the 2D pentagonal type PdQ₂ monolayers [17, 30-34]. Deng et al., [31] investigated the strain applied mechanical, electronic and optical properties of PdS₂, PdP₂ and PtSe₂. Except this, none of works have been reported on strain applied mechanical, electronic, optical and electronic transport properties of 2D penta-PdQ₂ (Q= S, Se). Further, the lower band gap of 2D-PdQ₂ monolayers has also motivated us to investigate absorbance of solar radiation on nanosheet. Hence, we also studied the strain dependent solar cell power conversion efficiency % (PCE) of penta-PdQ₂ (Q= S, Se) monolayers.

In this work, we have investigated the geometric, mechanical, electronic transport and optical properties of penta-PdQ₂ using density functional theory (DFT). After the optimization of structures, we analysed the lattice parameters (\AA), band gap (eV) and effective mass (m^*). Then after we applied biaxial strain ε (%) and obtained its bulk modulus B (GPa). The ultimate tensile strength (UTHS) is also found under the tensile strain ($+\varepsilon\%$) for PdQ₂ (Q= S, Se). We also studied the influence of loading on the band edges and obtained carrier mobility (μ_{2D}) along biaxial strain ε (%). We have adopted Shockley-Queisser (SQ) method to study Solar cell efficiency η (%) and calculated the maximum power density (P_{max}). The optical properties including the dielectric constants, absorption co-efficient, refractive index, and reflectivity of penta-PdQ₂ (Q= S, Se) monolayers are also reported in this paper. Herein, section 2 contains the methodology of computations, section 3 includes the results and discussion part of the work on penta-PdQ₂ (Q= S, Se) monolayers.

2. Computational Methods

The structural, electronic transport and optical properties of 2D wrinkled PdQ₂ (Q= S, Se) monolayers were performed with in the SIESTA code [35]. The exchange-correlation functional approach was used in term of Perdew–Burke-Ernzerhof (PBE) type pseudopotential [36]. The basic unit cell of 2D-PdQ₂ (Q= S, Se) contains two Pd atoms and four Q (Q=S, Se) atoms as shown in **Figure 1**. An energy cut-off of 450 and 300 Ry for PdS₂ and PdSe₂ monolayers were adopted, respectively. The doubled zeta plus (DZP) basis set was used with an energy of 0.02 Ry to expand the Kohn-Sham orbital. The Γ - centred mesh of $20 \times 20 \times 1$ and $15 \times 15 \times 1$ k-points were sampled for PdS₂ and PdSe₂ monolayers, respectively under the Monkhorst-pack scheme in the 2D Brillouin zone [37]. The force $0.01 eV/\text{\AA}$ was

kept to relaxing the unstrained and strained structures of penta-PdQ₂ (Q=S, Se) monolayers. The phonon dispersive curves were calculated using the density functional perturbation theory (DFPT) formalism [38]. In all calculation, the Fermi level (E_F) is shifted at the zero energy. The parameters of carrier mobility of penta-PdQ₂ (Q=S, Se) monolayers were found such as i.e., effective mass (m^*), stiffness constants (C_{2D}) and deformation energy (E_1) approximations using the following formula proposed by the Bardeen and Shockley [39], $\mu_{2D} = \frac{2e\hbar^3 C_{2D}}{3k_B T |m^*|^2 (E_1)^2}$; here, e is elementary charge of an electron, \hbar is the reduced Planck's constant, C_{2D} is the in-plane stiffness constant and for 2D system it is defined as $C_{2D} = \frac{1}{S_0} \frac{\partial^2 E}{\partial (\frac{a}{a_0})^2}$; where S_0 is cell area and a_0 is the lattice constant at equilibrium, E and a are the total energy and lattice constant of the monolayer after deformation, k_B is the Boltzmann constant, T is the temperature (300 K). Also, m^* is the effective mass in the transport direction and E_1 is the deformation potential (DP) constant denoting the shifting of each band edges due to the applied biaxial strain ε_{xy} defined as $E_1 = \frac{\partial E_{edge}}{\partial (\frac{a}{a_0})}$; where E_{edge} is energy value of VBM (for holes) and CBM (for electrons).

The electronic transport properties of PdQ₂ monolayers are calculated by utilizing the non-equilibrium Green's function (NEGF) in the form of I - V response by TRANSIESTA module of SIESTA code [40, 41]. The electric current through the scattering region is calculated by exploring the Landauer-Buttiker formalism,

$$I(V_b) = G_0 \int_{\mu_R}^{\mu_L} T(E, V_b) [f(E - \mu_L) - f(E - \mu_R)] dE \quad (1)$$

Where, $T(E, V_{bias})$ is the transmission probability of an electron incident at an energy E through the device under the bias voltage V_{bias} , μ_L and μ_R are the electrostatic potentials of left and right electrodes at a particular voltage bias and G_0 is the unit of quantum conductance. The optical properties were investigated including the dielectric function and absorbance coefficient by DFT within the random phase approximation (RPA) [42]. The dielectric function is given by $\varepsilon(\omega) = \varepsilon'(\omega) + i\varepsilon''(\omega)$. Here, $\varepsilon'(\omega)$ and $\varepsilon''(\omega)$ are the real and imaginary parts of the complex dielectric function [43], respectively.

3. Results and Discussion

3.1 Stability and Mechanical Properties

First, the geometric structures of pristine 2D penta-PdQ₂ (Q = S, Se) were systematically relaxed and lattice structures of penta-PdQ₂ were obtained. It is built up with two Pd and four Q (S, Se) atoms in the unitcell as shown in **Figure 1(a, b)**. The differential charge density in **Figure 1 (c, d)** shows an accumulation and depletion of charges between Pd and Q (S, Se) atoms. In **Table 1**, the relaxed structural parameters are summarized with ground state properties such as, lattice parameter (\AA), distance d (\AA) between Pd-Q₁, Pd-Q₂ and Q₁-Q₂, thickness t (\AA) and cohesive energy E_{coh} ($eV/atom$) of PdQ₂ monolayers.

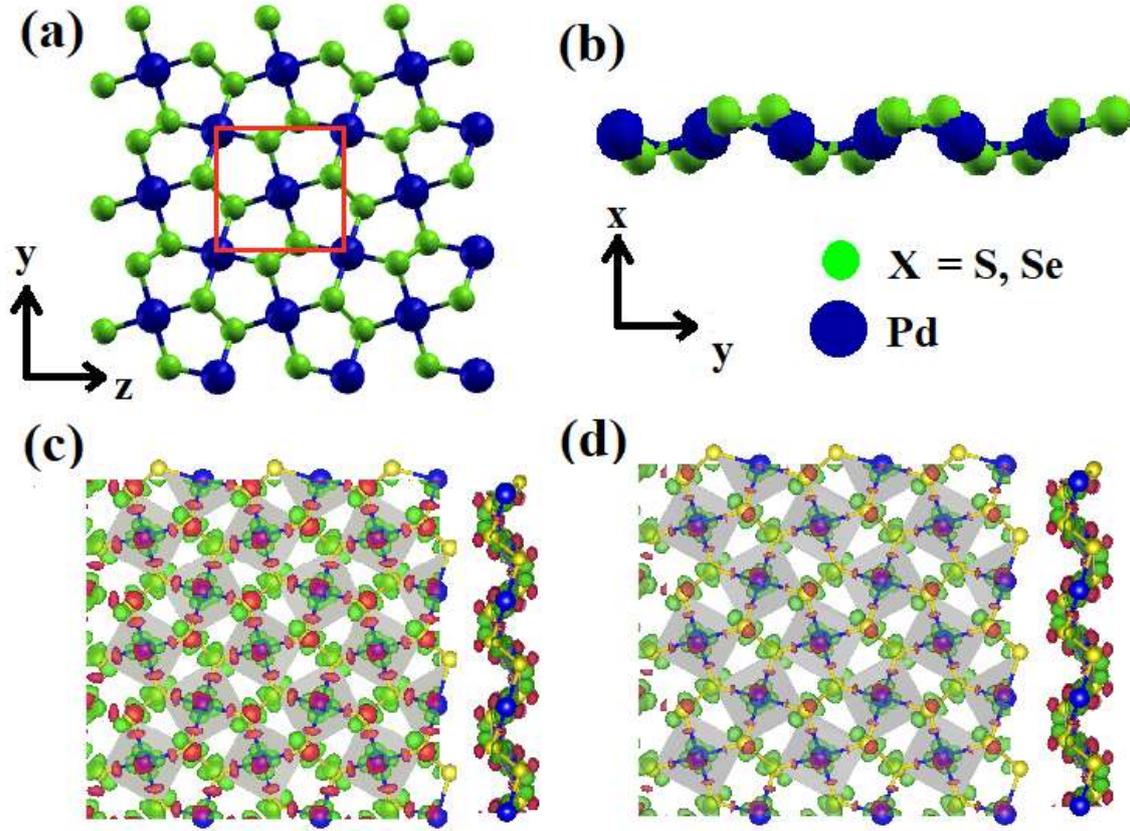


Figure 1. (Colour online) (a) Top-view of penta-PdQ₂ (Q = S, Se) monolayer structure. Red line indicates the unitcell of the system. (b) Side views from z - direction of the atomic structure of penta-PdQ₂. The navy blue and light-green circles represent the Pd and Q (S, Se) atoms, respectively. The deformation charge density plot (c) for penta-PdS₂ (d) for penta-PdSe₂; where red and green colour indicate the charge accumulation (electron excess) and charge depletion (electron loss). The iso-level is $0.063 e/\text{\AA}^3$ for both monolayers.

Table 1 : The calculated lattice parameter (\AA), bond lengths d (\AA), thickness t (\AA) and cohesive energy E_{coh} ($eV/atom$) of penta-PdQ₂ monolayers.

Penta-PdQ ₂ (Q = S, Se)		d _{Pd-Q1} (Å)	d _{Pd-Q2} (Å)	d _{X1-Q2} (Å)	Thickness (Å)	Lattice parameter (Å)	Cohesive energy <i>E_{coh}</i> (eV/atom)
Penta-PdS ₂	Present work	2.35	2.34	2.14	1.29	a= 5.62, b= 5.42	-4.71
	Other DFT/ *Exp.	2.34 [17] 2.35 [32]	2.35 [17] 2.34 [32]	2.10 [17] 2.11 [32]	1.28 [32]	a= 5.49, b = 5.59 [17] a= 5.48, b = 5.59 [32] a= 5.48, b = 5.57 [33]	--
Penta-PdSe ₂	present work	2.45	2.45	2.42	1.47	a= 5.94, b= 5.68	-4.30
	Other DFT/ *Exp.	2.45 [34]	2.46 [34]	2.42 [34]	--	a= 5.72, b= 5.80 [29] * a= 5.74, b= 5.91 [32] a= 5.75, b= 5.92 [33] a= 5.71, b= 5.90 [34]	--

As shown, in **Table 1**, the obtained lattice parameters a (b) of PdS₂ and PdSe₂ monolayers are 5.62 (5.42) Å and 5.94 (5.68) Å, respectively. The atomic distance between Pd-S₁(Se₁), Pd-S₂(Se₂), and S₁(Se₁)-S₂(Se₂) atoms are 2.35 (2.45) Å, 2.34 (2.42) Å and 2.14 (2.42) Å, respectively and shows good agreement with previously reported results [17, 29, 32-34]. To evaluate stability of resulted structures, the cohesive energy is calculated using the formula: $E_{coh} = \sum n_X E_X - E_{PdQ_2} / N$. where E_{PdQ_2} denotes total energy of PdQ₂ (Q = S, Se) unit cell, N is total atom in unit cell and n_X and E_X are number of atoms of each element and atomic energies of each atom in the unit cell, respectively. It is found that cohesive energies for PdS₂ and PdSe₂ monolayer are -4.70 and -4.30 eV/atom, respectively. Although this is larger than that for the 2D-antimony (-4.03), 2D-arsenene (-2.96), 2D-MoS₂ (-4.11), 2D-MoSe₂ (-3.94) and 2D-black phosphorous (-3.48). This is evidence that penta-PdQ₂ monolayers have strong bonding in its ring-network. In addition, to confirm the kinetic stability of penta-PdQ₂ (Q=S, Se) structures, we calculated the phonon dispersion curves along high symmetric points for both PdS₂ and PdSe₂ monolayers and results are as shown in **Figure S1 (a, b)**, ESI[†]. The highest phonon frequencies extended is up to 2275 cm^{-1} and 2193 cm^{-1} for PdS₂ and PdSe₂ monolayers, respectively. Evidently, no negative acoustics branch is appeared in the first Brillouin zone, itself proves the dynamical stability of both the monolayers at room temperature.

Next, we focused on mechanical properties of penta-PdQ₂ and evaluated Young modulus Y (N/m) and bulk modulus B (N/m). The Young moduli Y (N/m) and bulk moduli B

(N/m) were investigated under uniaxial and biaxial strain on the penta-PdQ₂ monolayers. The range of applied strain ε (%) is taken in interval of $-5\% \leq \varepsilon \leq +5\%$ in the step of 1%. The bulk modulus computed by applying biaxial load (x and y) on the penta-PdQ₂ monolayer with following relation, [44, 45]

$$B = S_0 \frac{\partial^2 E_s}{\partial A^2} \quad (2)$$

Where, S_0 is unstrained cross-sectional area of the unit cell. E_s and A are the total strain energy and area of respected applied strain, respectively. **Figure S2 (a, b), ESI[†]** shows the strain energy vs area curve for pristine and loaded structures of PdS₂ and PdSe₂ monolayers, respectively. This curve is fitted by the polynomial curve fitting and with help of Eq. (1), we have obtained bulk modulus B (N/m) as shown in **Figure S2 (a, b), ESI[†]**. The calculated bulk modulus (N/m) of PdS₂ and PdSe₂ are 30.22 and 23.56 N/m , respectively. These values are larger than that for the monolayer of Sb (~ 21.88 N/m) and As (~ 25.78 N/m) [44, 46], that indicate that penta-PdQ₂ has stiffer and better resistance to deformation compared to antimony (Sb) and arsenene (As) monolayers. Afterwards, we have calculated the in-plane Young modulus Y (N/m) in longitudinal (along x -axis) or transverse (along y -axis) direction for penta-PdQ₂. To obtained Young modulus from first-principles calculation, the following formula was employed [44, 45],

$$Y = \frac{1}{S_0} \frac{\partial^2 E_s}{\partial \varepsilon^2} \quad (3)$$

Here, S_0 is unstrained cross-sectional area of the unicell. Next, $\frac{\partial^2 E_s}{\partial \varepsilon^2}$ shows the second derivative of strain energy (eV) with applied load. Here, the compressive and tensile strains are applied to the penta-PdQ₂ in the longitudinal or transverse direction. The Young moduli obtained by using the curves shown in **Figure S3 (a, b), ESI[†]**. The calculated Young modulus Y (N/m) along x (y) direction are 74.13 (42.40) N/m and 65.26 (28.62) N/m for penta-PdS₂ and penta-PdSe₂, respectively. The results indicating that, Young modulus decreases as we switched S to Se, and this is due to the increment in the Pd-S and Pd-Se bond strength. Xiong et al. [47] had calculated orientation-dependent Young modulus of 2D orthorhombic MX₂ (M= Ni, Pd; X= S, Se, Te) and reported values are ~ 50 (37) N/m for PdS₂ (PdSe₂). From the present investigation of Young modulus of penta-PdQ₂, we suggest that the 2D penta-PdQ₂ can be preferred candidates for flexible devices because of having ultra-low values of Young modulus, compared to that of 2D graphene (340 N/m) and 2D MoS₂ (125 N/m) [48, 49].

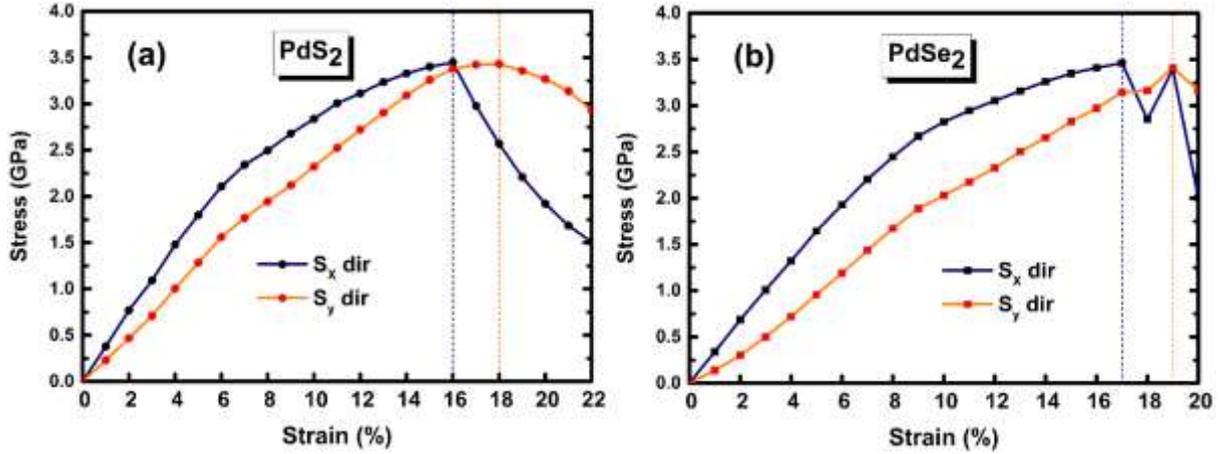


Figure 2. (Colour online) The stress (GPa) vs strain (%) curve under tensile strain. (a) For penta-PdS₂ and (b) For penta-PdSe₂.

Further, we have also explored the mechanical stability of penta-PdQ₂ monolayers and examined that up to which values of strain (%) the monolayers can withstand? It is worth to find curve of stress-strain relation called ultimate tensile strength (UTSH) curve [50]. The UTSH is representing the maximum stress value (GPa) that a monolayer can withstand prior to the fractured structure set. This can be calculated by the components of stress tensor with respect applied strain. The computed UTSH for penta-PdQ₂ monolayers are shown in **Figure 2 (a, b)**. The stress value, where the slope of the stress-strain curve becomes zero indicates the value of UTSH, and the strain at this value represents the magnitude of ultimate tensile strain (UTSR). In case of penta-PdS₂, the calculated values of UTSR along x (y) directions is 16% (18%) at stress of 3.44 GPa and 3.43 GPa, respectively (see **Figure 2(a)**). Whereas, in penta-PdSe₂ (see **Figure 2(b)**) it is observed at 17% (19%) with stress value of 3.46 GPa (3.40) GPa. The present values of UTSR for penta-PdQ₂ are higher than the blue-phosphorene monolayer (~16%) [51]. Thus, the UTSR values suggest that the penta-PdQ₂ monolayer is relatively more flexible.

3.2 Electronic Properties

The band structure, total density of states (TDOS) and partial density of states (PDOS) of penta-PdQ₂ monolayers along high symmetry points of Brillouin zone (BZ) have also investigated. At PBE level, from **Figure 3 (a, b)** we can see that the penta-PdS₂ (PdSe₂) is an indirect band gap with valence band maximum (VBM) and conduction band minimum (CBM) tracing at S- point (M-point) and Γ -point, respectively. The obtained indirect band

gap of penta-PdS₂ is $E_g^{\text{PBE}} = 0.94 \text{ eV}$, while band gap for penta-PdSe₂ is $E_g^{\text{PBE}} = 1.26 \text{ eV}$ as seen in **Figure 3 (a, b)**. We also did calculations using GGA+U, LDA, LDA+U and HSE06 approximations for PdS₂ (PdSe₂) and obtained indirect band gaps are 1.05 (1.26) eV, 0.92 (1.31) eV, 0.94 (1.30) and 0.92 eV, respectively. Next, having heavier element (Pd) in 2D penta-PdQ₂ material, the spin-orbit coupling (SOC) effect may play a crucial role and could influence in the band gap (E_g) of material. Thus, we have also checked the SOC effect on the penta-PdQ₂ monolayer and results are shown in **Figure S4 (a, b), ESI[†]**. Using PBE+SOC functional calculation, the obtained indirect band gap of penta-PdS₂ is $E_g^{\text{PBE+SOC}} = 1.16 \text{ eV}$, whereas band gap of penta-PdSe₂ is $E_g^{\text{PBE+SOC}} = 1.51 \text{ eV}$. Recently, the Yang et al [32], have reported electronic properties of penta-PdS₂ from PBE (PBE+SOC) calculation and reported indirect band gap of 1.08 eV (1.13 eV), which is comparable with our results within of 12.96% (11.5%) deviation. Moreover, a research group of Sun et al [34], have reported a band gap of penta-PdSe₂ with PBE level theory and mentioned indirect band gap of 1.38 eV, which is higher by 8.6% than our obtained band gap (~ 1.26 eV). Furthermore, with SOC effect, Qin et al [33], has computed indirect band gap of penta-PdSe₂ monolayer via TB-mBJ-GGA potential and reported band gap 1.38 eV. The deviation of present band gap of penta-PdSe₂ with PBE+SOC calculation is 9%. The presently obtained values are close to optimal value of 1.3 eV for solar cell applications. As presented in **Figure 3 (a, b)**, it is noticed that the TDOS (*States/eV*) are mostly ascend due to 'd' orbitals of Pd atom and 'p' orbitals of Q (S, Se) atom in the penta-PdQ₂ monolayer, which are also consistent to the past reported results [32, 34]. Additionally, the TDOS difference from PBE to PBE+SOC calculations are shown in **Figure S5 (a, b), (ESI[†])** for PdS₂ and PdSe₂ monolayers. we have also investigated PDOS (*States/eV*) as displayed in **Figure 3 (c, d)**, to further look into partial contribution of atoms in penta-PdQ₂ and we conclude that the conduction bands (CBs) and valence bands (VBs) are highly occupied by the *4d-Pd* states and *3p-Q* (Q =S, Se) states, respectively.

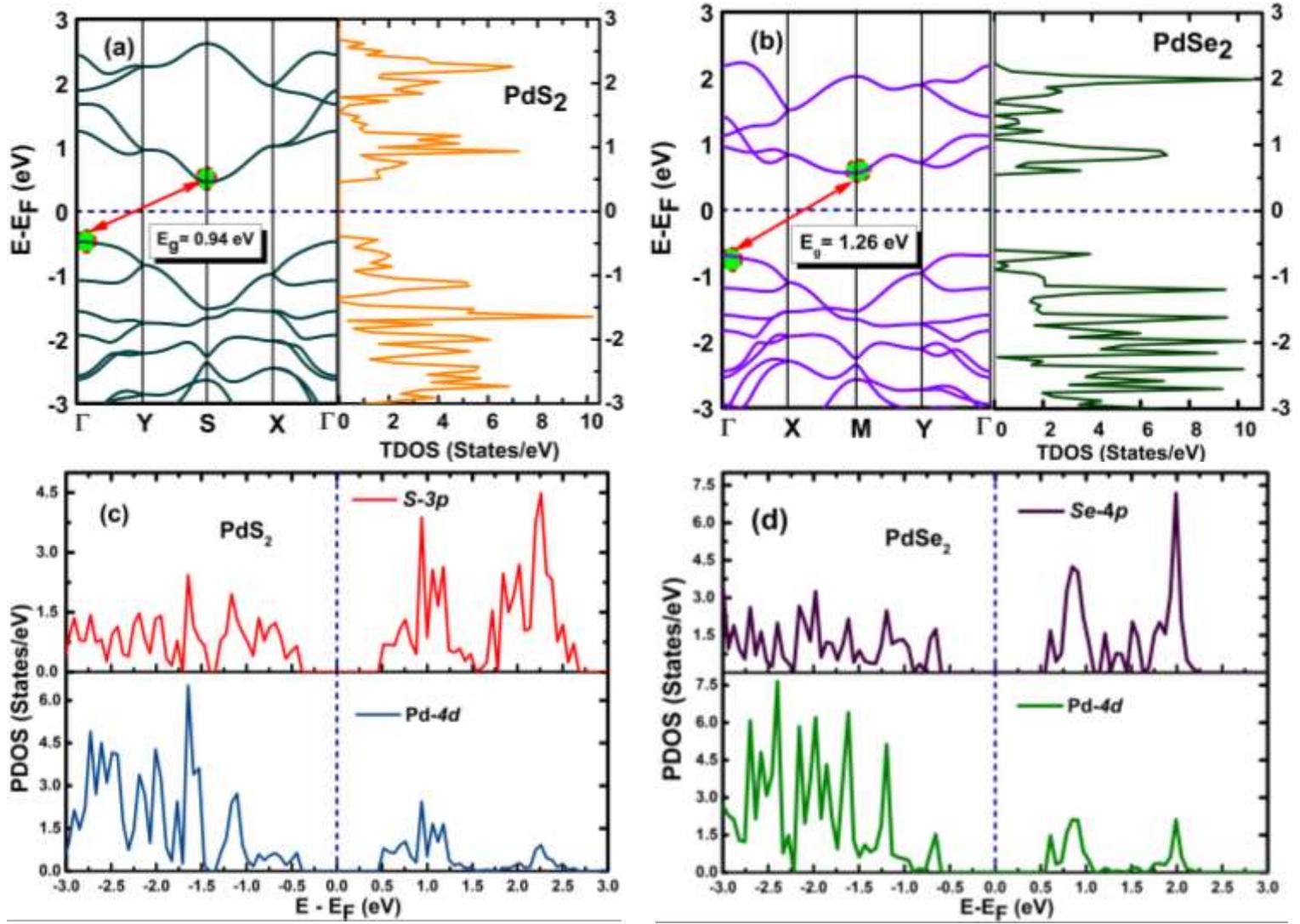


Figure 3. (Colour online) Electronic band structure and total density of states (TDOS) *States/eV* (a) For penta-PdS₂ monolayer and (b) For penta-PdSe₂ monolayer. The partial density of states (PDOS) *States/eV* (c) For penta-PdS₂ monolayer and (d) For penta-PdSe₂ monolayer.

3.3 Transport Properties

3.3.1 Carrier Mobility

We further focused on the band alignment in terms of strain effect to drive the electronic properties. Simultaneously, we have also calculated carrier mobility (μ) of monolayer, which is one of essential factor for a high-performance device like FET. For this, we have exerted compressive ($-\varepsilon\%$) and tensile ($+\varepsilon\%$) strain on penta-PdQ₂ (Q = S, Se) monolayers and traced

influence on the band edges of CBM and VBM in the band structure, where system is anisotropic (a not equal to b). The strength of strain along biaxial direction is given by [52], $\epsilon = [(S-S_0)/S_0] \times 100\%$; here, S_0 is the unstrained lattice constant and S is the strained lattice constant of the monolayer. The response of applied strain to band gaps of penta-PdQ₂ is shown in **Figure 4**.

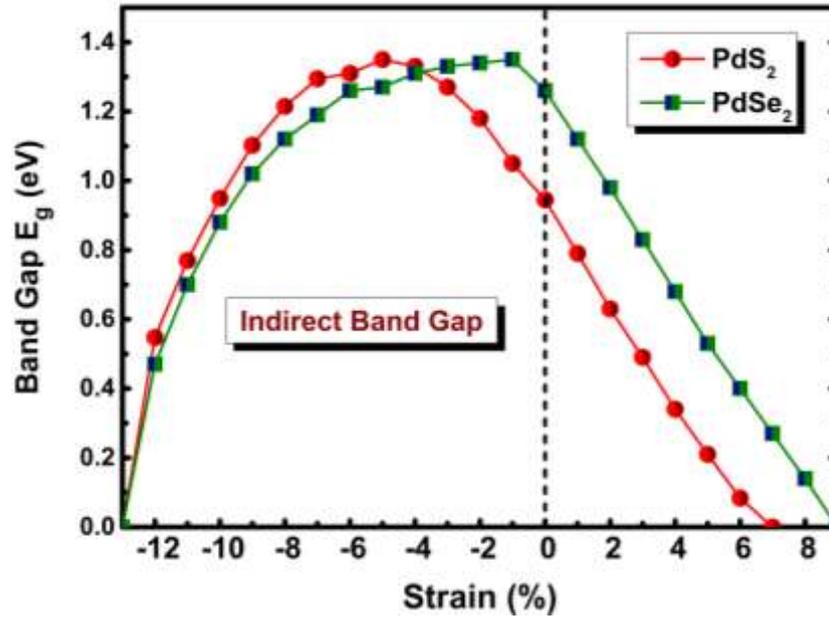


Figure 4. (Colour online) The variation of band gap E_g (eV) with applied biaxial strain ϵ (%) on penta-PdQ₂ monolayers.

As stated before, the fundamental indirect band gap of penta-PdS₂ and penta-PdSe₂ is 0.96 eV and 1.26 eV, respectively. The **Figure 4** reflects that the band gap of penta-PdQ₂ is gradually decreases almost linearly in presence of tensile strain ($+\epsilon\%$) compared to fundamental band gap (0%). Interestingly, upon increases compressive strain ($-\epsilon\%$) band gap of penta-PdS₂ increases up to -5% compressive strain and then decreases gradually with respect to 0%. It is noteworthy that, the maximum value of band gap of penta-PdS₂ is ~ 1.35 eV at $\epsilon = -5\%$ and for penta-PdSe₂ is ~ 1.36 eV at $\epsilon = -1\%$. Remarkably, the resistive phase transition is seen at higher compressive ($-\epsilon\%$) and tensile strain ($+\epsilon\%$) in both cases. The semiconducting nature of penta-PdS₂ (PdSe₂) is transform into semi-metallic (~ 0 eV) by applying the biaxial compressive and tensile strain of -13% (-13%) and +7% (+9%), respectively. Under the critical compressive strain ($-\epsilon\%$), the resistive phase change (semiconductor to semi-metallic) in penta-PdQ₂ behaviour arises due to crossing one band of VBM at the Fermi level (**Figure S6 (a, b), ESI[†]**). In contrary, up on the critical tensile ($+\epsilon\%$)

strain, it occurred due to touching one band of CBM at the Fermi level (**Figure S7 (a, b), ESI[†]**). Additionally, we have also examined the phonon spectrum for each applied strain on penta-PdQ₂ and computed its phonon frequency as summarized in **Table S1, ESI[†]**. Thus, our outcomes accomplish those electronic properties of studied 2D penta-PdQ₂ (Q= S, Se) materials are tuneable by applying biaxial strain, which establish that the penta-PdQ₂ is a promising candidate for designing flexible nanodevices.

Table 2. The carrier effective mass $|m^*|$ (m_e , the mass of free electrons), deformation

System	Carrier type	m^*/m_e	E_I (eV)	C_{2D} ($J m^{-2}$)	μ ($cm^2 v^{-1} s^{-1}$)	$R = \mu_h / \mu_e$
Penta-PdS ₂	electron	0.39	-6.44	121.76	258.06	0.26
	hole	0.72	6.85	121.76	67.43	
Penta-PdSe ₂	electron	0.58	-2.91	94.62	442.49	3.43
	hole	0.14	6.14	94.62	1518	

potential constant E_I (eV), stiffness constant C_{2D} ($J m^{-2}$), carrier mobility (μ) for electron (e) and hole (h) and carrier mobility ratio (R) along biaxial direction of penta-PdQ₂ monolayers at 300 K.

Owing to tunability of penta-PdQ₂ monolayers, we inspired to calculate the carrier mobility of penta-PdQ₂ monolayers. For that the Bardeen and Shockley formalism [39] had been applied, we have calculated the effective mass (m^*), deformation potential E_I (eV) and stiffness constant C_{2D} ($J m^{-2}$) to check the flow of charge carrier along biaxial strain. The carrier mobility depends on the most influenced factor on the carrier effective mass m^* of electron (e) and hole (h), that is directly derived from the electronic band structure as [53],

$$m^* = \hbar^2 \left\{ \frac{d^2E}{dk^2} \right\}^{-1} \quad (4)$$

Next, deformation potential E_I is another vital parameter to effect carrier mobility and is obtained from the deformation theory (DP) [39], which has been also successfully used method in previous reports for 2D MoS₂, WS₂ and phosphorene. The needed ingredients for carrier mobility (μ) are summarized in **Table 2**. Along biaxial strain, the obtained effective mass (m^*) of electron and hole for penta-PdS₂ (PdSe₂) are 0.39 m_e (0.58 m_e) and 0.72 m_e

($0.14 m_e$), respectively. Our computed deformation potential E_I (eV) for penta-PdS₂ is -6.44 eV for e and 6.84 eV for h , that is consistent with ones reported by Wang et al [17], along x or y direction. While in case of penta-PdSe₂, the value of E_I is -2.91 eV for e and 6.14 eV for h that is also in accordance with the results of Qin et al, [33] for x or y direction. Whereas the in-plane stiffness constant C_{2D} for penta-PdQ₂ has anisotropic behaviour. The in-plane stiffness constant obtained for penta-PdS₂ and penta-PdSe₂ are $121.76 J m^{-2}$ and $94.62 J m^{-2}$, respectively. The stiffness constant C_{2D} of penta-PdS₂ is larger than the penta-PdSe₂, indicating that PdSe₂ monolayer is softer than the penta-PdS₂ monolayer. As shown in **Table 2**, the achieved electron mobility of penta-PdS₂ monolayer is $258 cm^2 V^{-1} s^{-1}$, which is quite higher than the hole mobility $\sim 67 cm^2 V^{-1} s^{-1}$. Also, the carrier mobility ratio R comes out to be 0.26. Moreover, Wang et al [17] has reported the electron mobilities ($cm^2 V^{-1} s^{-1}$) of penta-PdS₂ as 40.97 (x) and 169.11 (y), while hole mobilities ($cm^2 V^{-1} s^{-1}$) as 339.25 (x) and 91.73 (y). On other hand, if we compare carrier transport along biaxial strain of penta-PdSe₂ the hole mobility is $1518 cm^2 V^{-1} s^{-1}$ that is greater than the electron mobility $\sim 442.49 cm^2 V^{-1} s^{-1}$ and R is 3.43. However, it is observed that the hole mobility of penta-PdSe₂ is higher than the 2D phosphorene [54] (i.e., $640-700 cm^2 V^{-1} s^{-1}$) and BN nanosheet (i.e., $500 cm^2 V^{-1} s^{-1}$) [55], indicating that penta-PdSe₂ would be a promising material for modelling electronic applications.

3.3.2 Current-Voltage (I-V) Characteristics

To understand the current sensitivity of the penta-PdQ₂ monolayers, we have examined the I-V characteristics, based on the equivalent transport theory [41]. **Figure 5** represents the schematic view of two-terminal device, where we have considered LE (left electrode), RE (right electrode) and scattering region of same material.

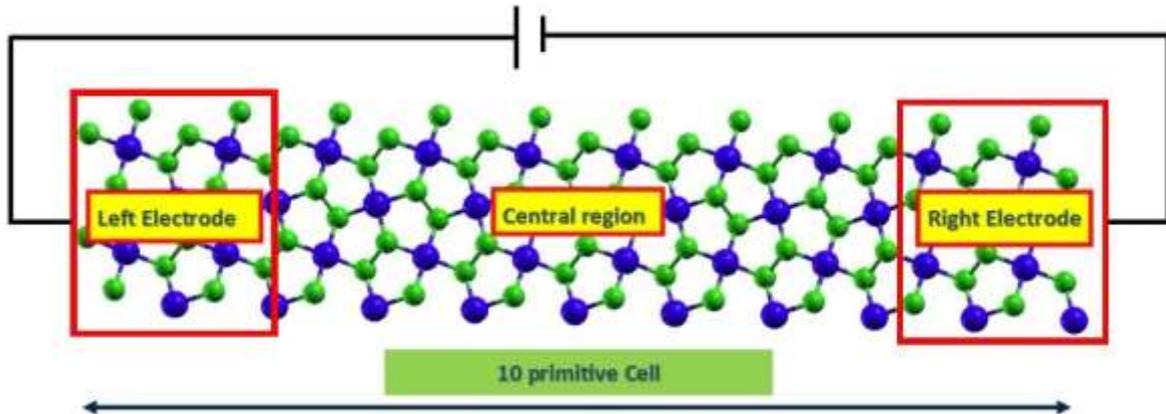


Figure 5. (Colour online) Schematic view of two-terminal device of penta-PdQ₂ monolayer. Here electrons are driven from the cold to the hot lead through central region.

Here, we have taken 10 primitive cells, among them the lead had built up with four-unit cell (24 atoms) and central region consist of 12-unit cells (72 atoms). The I-V characteristic of both penta-PdS₂ and penta-PdSe₂ monolayers are shown in **Figure 6 (a, b)**. It is seen that, no significant current is observed till ~ 1.0 V and 1.5 V then it starts to increase with increasing applied voltages due to semiconducting nature of penta-PdS₂ and PdSe₂ monolayers, respectively. Apparently, it can be seen from **Figure 6 (a, b)** penta-PdS₂ is more current-sensitive than the penta-PdSe₂ monolayer. Although, in the case of penta-PdS₂, the first peak in current is seen at 2.5 V with 14 μ A and second current peak is located at 4.0 V with current of 13.2 μ A. This is indicating a pronounced negative differential conductance effect (NDC) [56] in the bias range 2.0-3.5 V (see **Figure 6(a)**). While in case of penta-PdSe₂, the NDC effect is seen in range of 1.0-4.5 V (see **Figure 6(b)**). More precisely, the first NDC effect is occurs at the bias voltage 1.0 V and current to be found about 0.678 μ A. Further, the second and third peaks are occurring at the bias voltage of 3.0 V and 4.5 V with current up to 1.35 μ A and 2.05 μ A, respectively. The NDC effect is very useful feature in I-V curve because it would be playing a crucial role in the application of multipliers, mixers, logic gates, high-frequency oscillators, and A to D (analog-to-digital) converters [57].

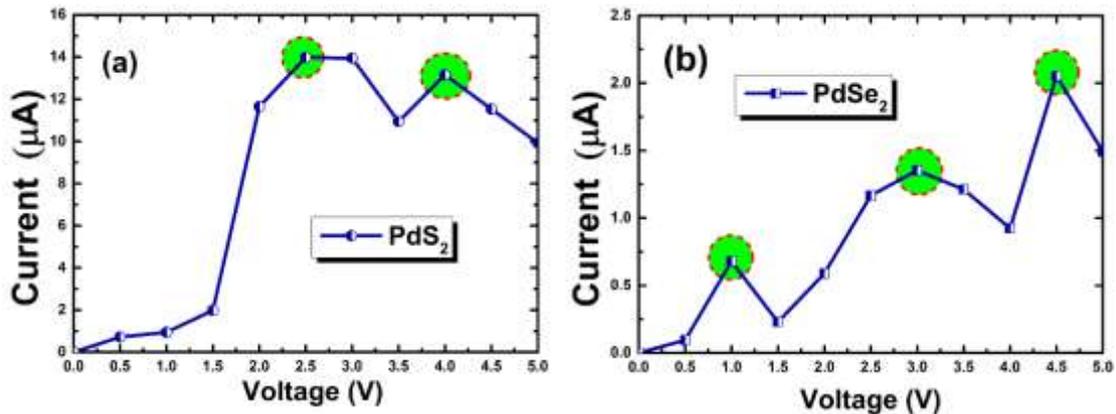


Figure 6. (Colour online) Current-voltage (*I-V*) plot of penta-PdQ₂ under bias voltages from 0.0 V to 5.0 V. The negative differential conductance (NDC) regions are indicated by the highlighted circles.

3.4 SQ Efficiency

We also focused on the power conversion efficiency (PCE) of penta-PdQ₂ (Q = S, Se) using Shokley-Queisser (SQ) limit that gives the percentage of power converted to electrical energy [58, 59]. For this we required several parameters such as short circuit current density (J_{sc}), open circuit voltage (V_{oc}), maximum power density (P_{max}) and efficiency η (%). The theoretical short circuit current density J_{sc} is calculated using following relation [60, 61],

$$J_{sc} = \int_0^{\infty} eA(E)I_{sun}(E)dE \quad (5)$$

where e is the elementary charge, $A(E)$ represents the absorbance of layer [60] and $I_{sun}(E)$ is the photon flux density as specified in the AM1.5G spectrum [62], Also, the theoretical reverse saturation current J_0 is defined as $J_0 = \int_0^{\infty} e\pi A(E)I_{bb}(E)dE$ [63]; Here $I_{bb}(E, T)$ is the black body spectrum and the radiative recombination fraction, which is supposed to be unity [58]. Thus, the total current density can be obtained using the formula [63],

$$J = J_{sc} - J_0 \left(\exp\left(\frac{eV}{kT}\right) - 1 \right) \quad (6)$$

where k is the Boltzmann's constant, V is the voltage over the absorber of the 2D devices. By the relation of $P = JV$, the maximum power density P_{max} can be evaluated from the maxima of the $J - V$ curve, as presented in **Figure 7(a, b)**. Eventually, with help of above-mentioned parameters, now we have obtained solar cell efficiency η (%) by formula [59],

$$\eta = \frac{P_{max}}{P_{in}} \quad (7)$$

where P_{in} (1000 W/m^2) is total incident power density from solar irradiation of AM1.5G and P_{max} is maximum power density.

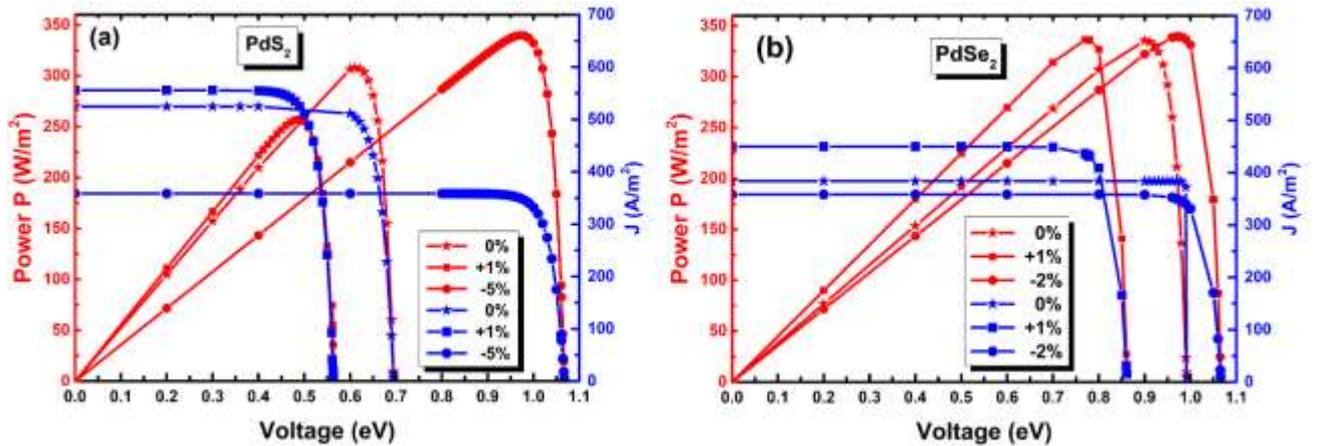


Figure 7. (Colour online) The calculated $J - V$ characteristic and power density curves for pristine and compressive ($-\varepsilon\%$) or tensile ($+\varepsilon\%$) loaded strain (maximum efficiency η ($\varepsilon\%$) of strain taken) at $T = 300\text{ K}$ (a) for penta- PdS_2 and (b) for penta- PdSe_2 .

As mentioned in the SQ limit theory, the electronic band gap (E_g) plays a pivotal role in the calculation of the material efficiency. Moreover, the maximum possible SQ efficiency limit is specified of 33.70% in circumstance of semiconductor with 1.34 eV optimum band gap [64]. As seen from the **Table 3**, the remarkable conversion efficiency for pristine penta- PdS_2 and penta- PdSe_2 is observed and are 30.79 and 33.54%, respectively. Also, the efficiency of penta- PdSe_2 is larger than the efficiency of benchmark GaAs ($\sim 31.4\%$) solar cell with single junction. Next, the band gap of both monolayers could be well tuned from 0.34 eV \sim 1.35 eV under influence of biaxial strain (%) as shown in **Figure 8 (a, b)**. Therefore, we have also checked the conversion efficiency under compressive ($-\varepsilon\%$) and tensile strain ($+\varepsilon\%$) for both the monolayers and presented in **Figure 8 (a, b)**. It is noted that, when we have exerted compressive strain ($-\varepsilon\%$) on the penta- PdQ_2 the efficiency at certain strain is higher as compared to the pristine (0%), that of 33.93% (at -5% of penta- PdS_2) and -33.94% (at -2% of penta- PdSe_2). On other hand, under tensile strain the maximum efficiency arises that of 25.72% at +1% of penta- PdS_2 and 33.63% at +1% of penta- PdSe_2 , which is lower than with respective each of pristine (0%). This suggests that, by applying biaxial strain on the penta- PdQ_2 the related SQ efficiency could be tuned. Also, the fabrications of penta- PdQ_2 solar cell have potential to improve photovoltaic performance compared to traditionally use Si-based solar cells.

Table 3. The calculated values of J_{sc} (A/m^2), P_{max} (W/m^2) and η (%) of penta- PdQ_2 (Q= S, Se) for both pristine and loaded strain (%).

2D-System	Strain (%)	J_{sc} (A/m^2)	P_{max} (W/m^2)	Efficiency η %
Penta-PdS_2	0 %	524.67	307.96	30.79 %
	+1 %	555.79	257.2	25.72 %
	-5 %	358.48	339.3	33.93 %
Penta-PdSe_2	0 %	383.82	335.4	33.54 %
	+1%	449.66	336.3	33.63 %
	-2%	358.48	339.48	33.94 %

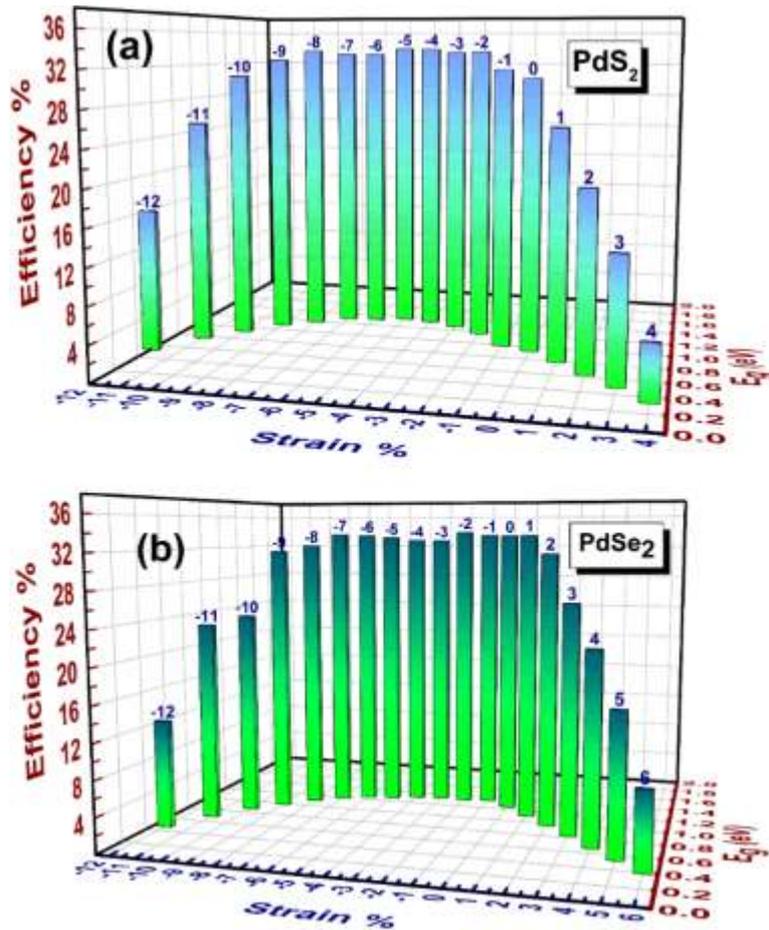


Figure 8. Change in SQ Efficiency η (%) of penta-PdQ₂ monolayers for pristine and loaded strain at T= 300 K (a) For penta-PdS₂ and (b) For penta-PdSe₂.

3.5 Optical Properties

In order to evaluate light absorption ability of penta-PdQ₂, we have calculated the real (ϵ') and imaginary (ϵ'') dielectric functions, absorption coefficient $I(\omega)$, refractive index $n(\omega)$, and reflectivity $R(\omega)$ of penta-PdQ₂ monolayers with parallel ($E \parallel c$) and perpendicular ($E \perp c$) electric vector (E). The computation is carried out using the Kramers-Kroning (KK) relationship [65] and results are shown in **Figures 9, S8 and S9, ESI†**.

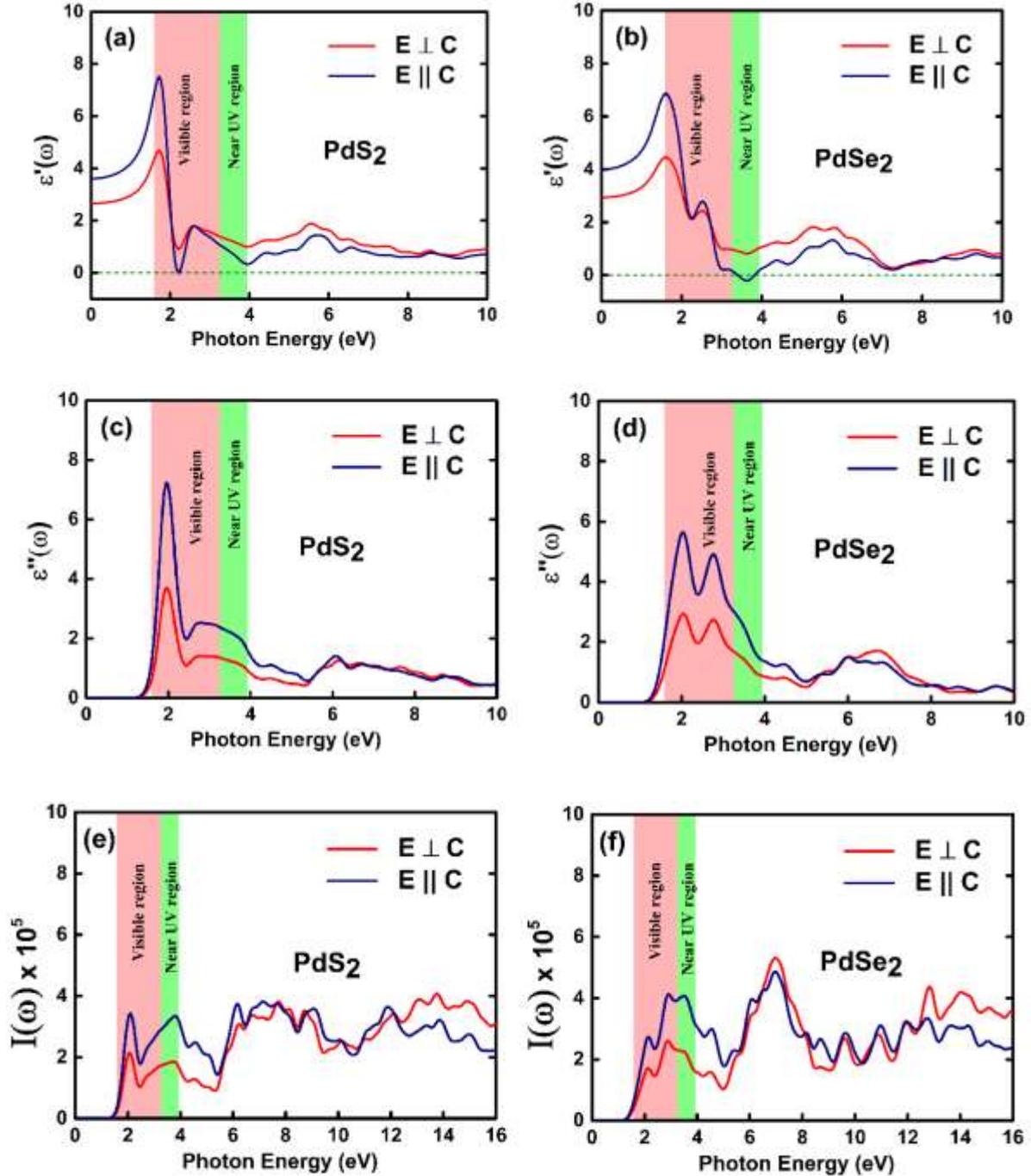


Figure 9. (Colour online) Computed optical properties of penta-PdQ₂ monolayers for parallel and perpendicular polarization (a, b) real (ϵ') and (c, d) imaginary (ϵ'') parts of complex dielectric function. (e, f) absorption coefficient $I(\omega)$.

In present work, the computed dielectric constants are shown in the large range of photon energy ($\hbar\omega$) from 0 to 10 eV and absorption coefficients $I(\omega)$ in 0 to 16 eV. The visible (VIS; 1.59 – 3.26 eV) and near UV (UV-A; 3.26 - 3.94 eV) spectrum regions are

shaded by the light-pink and light-green colour, respectively. **Figure 9 (a, b)** shows the graphical image of real (ϵ') and imaginary (ϵ'') components of penta-PdQ₂, that simply gives the idea about the electronic polarizability of the material from the Clausius - Mossotti [66] relation. It can be seen from the **Figure 9 (a, b)**, the static dielectric constant ($\epsilon_{\parallel}(0)$, $\epsilon_{\perp}(0)$) of penta-PdS₂ is (3.56, 2.62), while static dielectric constant of penta-PdSe₂ is 4.0 ($\epsilon_{\parallel}(0)$) and 2.94 ($\epsilon_{\perp}(0)$). This means that its $\epsilon(0)$ of parallel optical vector have more dominant polarizability compared to the perpendicular of $\epsilon(0)$. The optical peculiarity is strongly connected to the imaginary part of the dielectric function $\epsilon''(\omega)$, which reflects the transition between the energy bands near the E_F levels and that would be govern the linear response of the material to light under small wave vector as depicted in **Figure 9 (c, d)**. Therefore, the imaginary part $\epsilon''(\omega)$ in case of penta-PdS₂, the one major peak occurred at 1.98 eV in the visible (VIS) region for both $\epsilon''_{\perp}(\omega)$ and $\epsilon''_{\parallel}(\omega)$ case as plotted in **Figure 9 (c)**. While, in case of penta-PdSe₂, it is clearly seen (**Figure 9 (d)**) that there are two intense peaks appeared in the visible region (VIS) at $\hbar\omega = 2.04$ eV and 2.76 eV due to the electronic transition of '4p' states of Se atom and '4d' states of Pd atom between conduction bands (CBs) and valence bands (VBs) of the electronic band structure. Apparently, the peaks tendency of $\epsilon''_{\perp}(\omega)$ and $\epsilon''_{\parallel}(\omega)$ are similar on the photon energy spectrum for both penta-PdQ₂ monolayers.

The optical absorption spectra of the penta-PdQ₂ were recorded by absorption coefficient $I(\omega)$ vs photon energy (eV) as shown in **Figure 9 (e, f)**. For penta-PdQ₂, the first absorption peak orientation is in the ~ 1.5 -5 eV and second broad peak's orientation is cover the UV region up to photon energy of 16 eV. For penta-PdS₂, the low energy excitonic peak occur around 2.1 eV in infrared region (IR) and that peak rise towards the visible region at 3.78 eV for both the polarization. The maximum absorption coefficient $I(\omega)$ of penta-PdS₂ for parallel and perpendicular polarization is 3.65×10^5 and 4.04×10^5 , respectively (See **Figure 9 (e)**). While, in case of penta-PdSe₂, the first peak is located at 2.2 eV in the IR region and kick off in visible region at highest absorption coefficient $I(\omega)$ of 4.12×10^5 . However, with perpendicular polarization the highest coefficient found is about 5.28×10^5 in the UV region (See **Figure 9 (f)**). Thus, the penta-PdQ₂ monolayers have shown prominent absorption in VIS and UV region, which suggest a good prospect of penta-PdQ₂ monolayers as UV filters and UV photodetectors.

Next, the computed refractive index $n(\omega)$ and reflectance $R(\omega)$ of penta-PdQ₂ have the same evolution curve as plotted in **Figure S8 (a, b), ESI[†] and S9 (a, b), ESI[†]**. In

penta-PdS₂, we observed static refractive index as $(n^{\parallel}(0), n^{\perp}(0)) = (1.90, 1.63)$ (see **Figure S8 (a), ESI[†]**). With the parallel polarization, the $n(\omega)$ is rising in the VIS region up to $\hbar\omega = 1.78$ eV, then going down up to energy of 2.32 eV. Similarly, in case of perpendicular polarization it has also gone down in the VIS region with 1.23 of $n(\omega)$ by the same spectrum photon energy. Moreover, the reflectivity $R(\omega)$ is also shown in **Figure S9 (a), ESI[†]**. The static reflectance $(R_{\parallel}(0), R_{\perp}(0))$ of penta-PdS₂ in VIS region is about to (10%, 5%) and the most elevated peak of reflectance is found to be in the VIS region with reflectance of 31% as seen in **Figure S9 (a, b), ESI[†]**. While, for penta-PdSe₂, the static refractive index $(n^{\parallel}(0), n^{\perp}(0))$ is (2.0, 1.71) as shown in **Figure S8 (b), ESI[†]**, which is also in good agreement with previous reported work on penta-PdSe₂ monolayer by the Zhao et al [67] group. The first and highest peak of $n(\omega)$ is trapped in the VIS region at energy of 1.68 eV, at that energy the $n^{\parallel}(\omega)$ is 2.65. The intense peak of $n^{\perp}(\omega)$ is also located at $\hbar\omega = 1.68$ eV in the VIS region with $n^{\perp}(\omega)$ of 1.68. The static reflectance $(R_{\parallel}(0), R_{\perp}(0))$ of penta-PdSe₂ in VIS region is about to (11%, 7%) as seen in **Figure S9 (b), ESI[†]**. Also, the highest peak of $R_{\perp}(\omega)$ is occurred in the VIS region with the reflectance of 26%, that reflectance is in good agreement with Zhao et al [67] reported results of penta-PdSe₂ monolayer. Overall, the prominent peaks appeared in the $n(\omega)$ and $R(\omega)$ in the VIS region, that means most of incident light energy is reflected and refracted in VIS region and only small amount of incident energy is absorbed by the material.

4. Conclusions

In the summary of the work, the first principles method has been employed to study the band structure, bulk modulus, Young's modulus, transport properties such as carrier mobility and I - V curve, and optical properties of penta-PdQ₂ (Q= S, Se) monolayers. The positive phonon spectrum gives the ground state dynamical stability of both the materials. The obtained electronic band gap is 0.94 eV and 1.26 eV for penta-PdS₂ and penta-PdSe₂, respectively. From mechanical stability point of view, penta-PdS₂ withstands up to 16% (18%) in x (y) directions, while penta-PdSe₂ is up to 17% (19%), which undoubtedly implies that each monolayer is flexible up to that strain. Interestingly, the I - V curve shows the NDC effect beyond the bias voltage of 2.5 V (for PdS₂) and 3.0 V (for PdSe₂), hence this feature leads us to conclude that penta-PdQ₂ monolayers will be consider in future as promising material for NDC-based nanodevices. More importantly, the SQ efficiencies for pristine PdS₂ and PdSe₂ were 30.79% and 33.54%, respectively, that could be essential utilized in the solar cell application. The computed optical properties reveal that the absorption range of penta-PdQ₂

is very broad in *UV* regions. The maximum absorption coefficient $I(\omega)$ of penta-PdS₂ and penta-PdSe₂ are found in the *UV* region are 4.04×10^5 and 5.28×10^5 , respectively. Collectively, due to unique electronic, mechanical, transport and optical properties of penta-PdQ₂ monolayers, the 2D materials have application prospect in the arena of semiconducting nanodevices.

Conflicts of interest

The authors declare that they have no competing interests.

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Author Contributions

DR, S.K.G and P.N.G studied conception and designed the final approval of the version to be published. DR was associated with the DFT calculations. S.K.G and P.N.G helped to improve the scientific content of the article. All author reviewed the manuscript.

Competing Interests

The authors declare no competing interest.

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