

Exploring the anodic performance of InSnPS/GaGeAsSe heterostructure as an ultra-high rate rechargeable Li ion battery via first-principles study

Francis Opoku^{1,2,*}, Eric Selorm Agorku¹, Michael Kumi³, and Penny Poomani Govender^{2,†}

¹Department of Chemistry, Kwame Nkrumah University of Science and Technology, Kumasi, Ghana

²Department of Chemical Sciences, Doornfontein Campus, University of Johannesburg, P.O. Box 17011, Johannesburg 2028, South Africa

³CSIR-Water Research Institute, Environmental Chemistry and Sanitation Engineering Division, P.O. Box AH 38, Achimota-Accra, Ghana



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Because heterostructures have a layered structure, a large surface area, and several accommodating sites, they efficiently maximize the characteristics of rechargeable Li-ion batteries (LIBs). Based on density functional theory calculations, the electrochemical, electronic, diffusion, adsorption, and mechanical properties of InSnPS/GaGeAsSe heterostructure as anode material for LIB are investigated in this work. The predicted 2D heterostructure has a mechanically and dynamically stable structure with a small lattice mismatch (2.92%). The InSnPS/GaGeAsSe effectively isolates the electrons and holes at the interface by creating a staggered type II band alignment, extending the carrier lifetime. Additionally, the InSnPS/GaGeAsSe heterostructure has low average open circuit voltages of 0.69 V and a storage capacity of 483.38 mAh g⁻¹ because of its energetically advantageous adsorption of five layers of Li atoms. The remarkable charge-discharge capabilities of the InSnPS/GaGeAsSe are demonstrated by the low activation barrier of 0.23 eV, which illustrates the quicker mobility of Li ions. Considering the high theoretical specific capacities, low diffusion energy barriers, strong adsorption properties, structural integrity and low open circuit voltages, the InSnPS/GaGeAsSe heterostructure is predicted to be the preferable anode material for LIBs.

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I. INTRODUCTION

The world's population continues to grow, and the technology era is developing quickly, increasing energy demand [1]. Rechargeable, cutting-edge, reasonably priced, lightweight, and long-lasting batteries are needed to meet the growing demand for electric vehicles and portable electronics [2]. Batteries are essential for energy storage systems due to their high energy density, compact size, and superior operating stability [3,4]. Intelligent electronic devices, electric cars, and smart portable electronics like drones and cell phones all employ Li-ion batteries (LIBs), one of the most popular commercialized batteries [5,6]. Because of its exceptional performance, which includes a low self-discharge, no memory effect, and high energy density [7], LIBs are effective and reversible energy storage devices with a long cycle life, a wide voltage window, and high energy density [8,9].

Because of its Coulomb efficiency and high cycle stability, graphite is being used as the anode material for LIBs [5]. Nonetheless, its high diffusion energy barrier of 0.45–1.20 eV and low specific capacity of 372 mAh g⁻¹ restrict its potential uses [10]. As such, it does not meet the minimum

standards needed for long-range electric vehicles. To boost the performance of LIBs, new suitable anode materials with high charge/discharge efficiency, more space for ion loading and tiny volume variations are urgently required [11]. Several two-dimensional (2D) materials have lately been examined as anode materials in metal-ion batteries because of their fast charge/discharge rates, high energy densities, specific surface area, electrical conductivity, and stability [12–14]. Several 2D materials based on III–V and V–VI group elements have recently been explored for their promising electrochemical properties in LIBs. For example, theoretical investigations have shown that monolayer InP₃ exhibits favorable metallic conductivity upon lithiation, strong Li binding, and low diffusion barriers, suggesting its potential as a high-rate anode material [15]. Similarly, 2D GaN has been predicted to offer exceptional Li storage capacity and ultrafast ion diffusion, with theoretical values far surpassing conventional anode materials [16]. Experimental realization of 2D GaN has also been demonstrated, confirming the feasibility of synthesizing group III–V layered structures under appropriate conditions [17]. Additional studies on arsenene and β -In₂Se₃ monolayers have further shown that group-V-based 2D materials can exhibit both high lithium storage capacity and low activation energy for Li diffusion [18,19]. Moreover, layered In₂Se₃ and InSe have been synthesized and evaluated as LIB anodes, showing excellent cycling stability and near-unity Coulombic efficiency across hundreds of charge-discharge cycles [20]. The functionalized MBene TiBX (X = Si, Ge, P, As) structures exhibit high theoretical specific capacities, low open-circuit voltages, and low diffusion barriers, highlighting their potential as promising electrode materials

*Contact author: ofrancis2010@gmail.com;
francisopoku@knust.edu.gh

†Contact author: pennyg@uj.ac.za

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[21]. The $\text{Ga}_3\text{C}_6\text{N}_6$ monolayer demonstrates a well-rounded performance, characterized by robust adsorption energies, low diffusion barriers, favorable average open-circuit voltages, and high theoretical specific capacities [22].

The open channels of 2D material allow ion transport and make its surfaces accessible, resulting in increased capacity and rapid charge storage compared to their bulk counterparts. However, due to the possibility of irreversible interactions at the solid/electrolyte interface brought on by its huge active surface area, a single 2D material can scarcely satisfy the numerous requirements for high-performance anode materials. Building a van der Waals heterostructure (vdWH) is one of the most popular approaches to maximize the qualities of 2D materials as anode materials for rechargeable LIBs and combine the advantages of two distinct components. For example, Bediako *et al.* observed improved Li^+ transfer capability and high carrier density doping through theoretical calculations and experimental investigations when a graphene/ MoX_2 heterostructure was used as anode material for LIBs [23]. Li *et al.* [24] used DFT simulations to predict that $\text{MoS}_2/\text{Ti}_2\text{CT}_2$ ($T = \text{O}, \text{F}$) heterostructure will make ideal anode materials for LIBs owing to their low diffusion barriers (0.57 eV), high theoretical capacity, and large tensile strains (>20%). The high capacities, low open circuit voltages (OCV), suitable diffusion energy barriers, and strong adsorption properties of $\text{VS}_2/\text{Ti}_2\text{CT}_2$ ($T = \text{O}, \text{S}$) heterostructure were designed to be promising high-performance anode materials for LIBs [25]. The $\text{MoS}_2/\text{M}_2\text{CS}_2$ ($M = \text{Ti}, \text{V}$) heterostructure exhibits metallic conductivity, structural stability, and low diffusion barriers (0.12–0.38 eV) due to the S-functionalized M_2CS_2 layer, indicating excellent rate performance for LIB applications [26].

We explore the design of novel Janus materials composed of Group III–V elements as promising anode candidates for LIBs. Although recent progress has been made in developing III–V heterostructure anodes, this study provides a first-principles investigation into the previously unexamined $\text{InSnPS}/\text{GaGeAsSe}$ system. The Janus InSnPS and GaGeAsSe monolayers are formed by disrupting out-of-plane symmetry, resulting in a unique structural configuration. Due to their comparable hexagonal frameworks and closely matched lattice constants, these monolayers are expected to form a stable heterostructure with minimal lattice strain. This work evaluates the potential of the $\text{InSnPS}/\text{GaGeAsSe}$ heterostructure as an efficient LIB anode material by analysing its electronic, structural, and mechanical properties and electrochemical behavior. Key performance metrics, such as open-circuit voltage (OCV), theoretical capacity, and lithium adsorption and diffusion characteristics, are used to assess its suitability. The findings provide theoretical insights and guidance for developing high-performance anode materials for rechargeable ion batteries.

II. COMPUTATIONAL DETAILS

Using QUANTUM ESPRESSO [27,28], density functional theory simulations were performed to examine the properties of $\text{InSnPS}/\text{GaGeAsSe}$ heterostructure. The optimized norm-conserving pseudopotentials described the electron-ion

interaction with plane-wave cut-off energy of 50 Ry [29]. The selected valence-electron configurations are In ($5s^25p^1$), Sn ($5s^25p^2$), P ($3s^23p^3$), S ($3s^23p^4$), Ga ($4s^24p^1$), Ge ($4s^24p^2$), As ($4s^24p^3$), and Se ($4s^24p^4$). Grimme's DFT–D3 method [30,31] with Becke–Jonson damping dispersion corrections was used to account for the vdW interactions, while the electron exchange–correlation potentials were described using the Perdew–Burke–Ernzerhof (PBE) functional within the general gradient approximation [32]. During geometry optimization, Monkhorst–Pack mesh with $4 \times 4 \times 1$ and $12 \times 12 \times 1$ k-point was used to sample the Brillouin zone of the 3×3 supercell and unit cell, respectively. Because of the asymmetric layer design, dipole correction [33] was considered for all calculations. Li atom adsorption and diffusion behaviors on $\text{InSnPS}/\text{GaGeAsSe}$ are calculated using a 3×3 supercell with a vacuum layer of 30 Å. All structures were fully optimized with the convergence thresholds for energy and force set to less than 10^{-8} Ry and 10^{-3} Ry Bohr^{-1} , respectively. Methfessel–Paxton smearing with $\text{degauss} = 0.02$ Ry was adopted to ensure smooth convergence. To separate the two layers of the heterostructure, the thickness of the vacuum layer was adjusted to 30 Å to remove periodic image interaction along the z -axis. Utilizing the finite displacement method [34], the phonon spectra are computed using a cutoff radius of 19 Å and a 5×5 supercell for the $\text{InSnPS}/\text{GaGeAsSe}$ heterostructure and a 6×6 supercell for the InSnPS and GaGeAsSe monolayers. Moreover, the HSE06 hybrid functional [36] was chosen for computing the electronic properties because the PBE functional underestimates the bandgap of materials. The diffusion energy barrier of Li atoms on the $\text{InSnPS}/\text{GaGeAsSe}$ surface was determined via the climbing image nudged elastic band (CI–NEB) method [37] with a seven-image pathway. Using Bader charge analysis [38,39], the charge transfer was quantitatively investigated.

Cohesion energy (E_{coh}) is used to assess the possibility of the experimental fabrication of 2D materials [40]. It is computed using the following equation:

$$E_{\text{coh}} = \frac{(nE_{\text{Ga}} + nE_{\text{Ge}} + nE_{\text{As}} + nE_{\text{Se}} - E_{\text{GaGeAsSe}})}{4} \quad (1)$$

where E_{Ga} , E_{Ge} , E_{As} , and E_{Se} signify the energy of Ga, Ge, As, and Se atoms, respectively, and the E_{GaGeAsSe} signifies the total energy of the GaGeAsSe . n is the number of each atom.

$$E_{\text{coh}} = \frac{(nE_{\text{In}} + nE_{\text{Sn}} + nE_{\text{P}} + nE_{\text{S}} - E_{\text{InSnPS}})}{4} \quad (2)$$

where E_{In} , E_{Sn} , E_{P} , and E_{S} denote the energy of In, Sn, P, and S atoms, respectively, and the E_{InSnPS} denotes the total energy of the InSnPS .

The interface binding energy of the heterostructure is defined as [26]

$$E_b = (E_{\text{InSnPS}/\text{GaGeAsSe}} - E_{\text{InSnPS}} - E_{\text{GaGeAsSe}})/A \quad (3)$$

where $E_{\text{InSnPS}/\text{GaGeAsSe}}$, E_{InSnPS} , E_{GaGeAsSe} , and A are the total energies of the $\text{InSnPS}/\text{GaGeAsSe}$, the InSnPS , and GaGeAsSe , respectively, and A is the interface surface area.

For ion adsorption, the adsorption energy (E_{ad}) between Li and $\text{InSnPS}/\text{GaGeAsSe}$ heterostructure is obtained by the

TABLE I. Calculated lattice constant ($a = b$), interlayer distance (d), bonding length, work function (Φ), charge transfer ($\Delta\rho$), interface binding energy (E_b), and band gap (E_g) for the monolayers InSnPS and GaGeAsSe and the stacking patterns of InSnPS/GaGeAsSe heterostructure.

System	$a = b$ (Å)	d (Å)	$d_{\text{In-S}}$ (Å)	$d_{\text{In-Sn}}$ (Å)	$d_{\text{Sn-P}}$ (Å)	$d_{\text{Ga-Se}}$ (Å)	$d_{\text{Ga-Ge}}$ (Å)	$d_{\text{Ge-As}}$ (Å)	Φ (eV)	$\Delta\rho e $	E_b (eV/Å ²)	E_g (eV)
InSnPS	3.922		2.547	2.858	2.549				5.81			2.39
GaGeAsSe	3.809					2.491	2.472	2.482	5.15			2.50
A1	3.866	3.737	2.533	2.828	2.532	2.504	2.458	2.498	4.71	0.2187	-15.345	1.96
A2	3.866	3.681	2.533	2.824	2.531	2.503	2.457	2.497	4.85	0.2283	-15.506	1.91
A3	3.866	3.726	2.533	2.825	2.531	2.511	2.464	2.496	4.84	0.2112	-14.665	2.03
A4	3.866	3.674	2.533	2.829	2.532	2.503	2.457	2.498	4.54	0.2403	-15.761	1.71

following equation [41]:

$$E_{\text{ad}} = \frac{E_{(\text{InSnPS/GaGeAsSe}+n\text{Li})} - E_{(\text{InSnPS/GaGeAsSe})} - nE_{\text{Li}}}{n} \quad (4)$$

where $E_{(\text{InSnPS/GaGeAsSe}+x\text{Li})}$, $E_{(\text{InSnPS/GaGeAsSe})}$, and nE_{Li} signify the total energies of the Li/InSnPS/GaGeAsSe, InSnPS/GaGeAsSe, and isolated Li atom, respectively, and n represents the total amount of the adsorbed Li ions in InSnPS/GaGeAsSe heterostructure with a 3×3 supercell. A negative E_{ad} indicates that the Li atom can be stable adsorbed on the surface of the heterostructure.

The theoretical storage capacity of InSnPS/GaGeAsSe heterostructure is evaluated as [42]

$$C = \frac{znF}{M_{\text{InSnPS/GaGeAsSe}}} \quad (5)$$

where z , n , F , and $M_{\text{InSnPS/GaGeAsSe}}$ are the valence of Li (1 for the alkali metal ions), number of Li atoms adsorbed per unit of InSnPS/GaGeAsSe, Faraday constant (26 801 mA h mol⁻¹), and the molecular weight of InSnPS/GaGeAsSe heterostructure, respectively.

In the process of electrochemical reaction, the OCV of Li on InSnPS/GaGeAsSe is calculated via [42]

$$\text{OCV} = \frac{E_{(\text{InSnPS/GaGeAsSe})} + n\mu_{\text{Li}} - E_{(\text{InSnPS/GaGeAsSe} + n\text{Li})}}{ne} \quad (6)$$

where n is the amount of Li atoms adsorbed on InSnPS/GaGeAsSe and e is the elementary charge.

For 2D materials, mechanical properties such as elastic constants, Young's modulus and Poisson's ratio were calculated using thermo_pw.x [43] with the elastic tensor derived from the stress-strain relationship and total energy methods as follows [44]:

$$\nu = \frac{C_{12}}{C_{11}} \quad (7)$$

$$Y = \frac{C_{11}^2 - C_{12}^2}{C_{11}} \quad (8)$$

where C_{11} and C_{12} are the elastic constants. Small deformations (0.5%) and a 2D layer thickness of 3.35 Å were used to compute the stress-strain relationship. Postprocessing and visualization of angle-dependent Young's modulus, Poisson's ratio, and other mechanical properties were performed using ELATools [45].

III. RESULTS AND DISCUSSION

The structural and electronic properties of InSnPS and GaGeAsSe monolayers are first examined to produce the stable InSnPS/GaGeAsSe heterostructure (see Table I). The Bravais vectors of optimized InSnPS monolayer [Fig. 1(a)] are $a = b = 3.922$ Å. The In-S, In-Sn, and Sn-P bond lengths are 2.547, 2.858, and 2.549 Å and the S-In-Sn and In-Sn-P bond angles are around 117.230° and 117.324°. The optimized structure of the GaGeAsSe monolayer with a hexagonal cell is shown in Fig. 1(b), and the lattice constant is $a = b = 3.809$ Å.

The calculated in-plane bond lengths for Ga-Se, Ga-Ge, and Ge-As bonds are around 2.491, 2.472, and 2.482 Å, respectively. As depicted in Figs. 1(c) and 1(d), the InSnPS and GaGeAsSe monolayers show an indirect band gap value of 2.39 and 2.50 eV, respectively. The valence band maximum (VBM) is situated between Γ and M symmetry points. Concurrently, the Γ and M symmetry points correspond to the conduction band minimum (CBM) of InSnPS and GaGeAsSe monolayers, respectively. The P 3*p* and S 3*p* orbitals contribute to both the VBM and CBM of the InSnPS monolayer [Fig. 1(e)]. In the meantime, the 4*p* orbitals of As and Se primarily contribute to the CBM and VBM of the GaGeAsSe monolayer [Fig. 1(f)].

A unit cell of InSnPS monolayer is stacked with a unit cell of GaGeAsSe monolayer along the normal direction to create InSnPS/GaGeAsSe vdWH. Heterostructures are widely thought to be possible when the lattice mismatch is less than 5%, which is calculated as follows [46]:

$$\delta = \frac{2|a_1 - a_2|}{a_1 + a_2} \times 100\%, \quad (9)$$

where a_1 and a_2 denote the lattice constants of InSnPS and GaGeAsSe monolayers. The lattice mismatch of 2.92% is very beneficial for experimental synthesis [47]. Scalable synthesis of 2D heterostructures can be achieved via advanced chemical vapor deposition (CVD) and epitaxial techniques. For instance, chemical vapor deposition has enabled the controllable bottom-up growth of high-quality vertical heterostructures, particularly in transition-metal dichalcogenides [48]. CVD and epitaxial techniques offer viable routes for scalable synthesis of such 2D layers [49]. Additionally, epitaxial approaches, both van der Waals and physical, have produced wafer-scale, single-crystal films of transition metal dichalcogenide suited for heterostructure integration [50]. Notably, van der Waals heteroepitaxy using a two-step

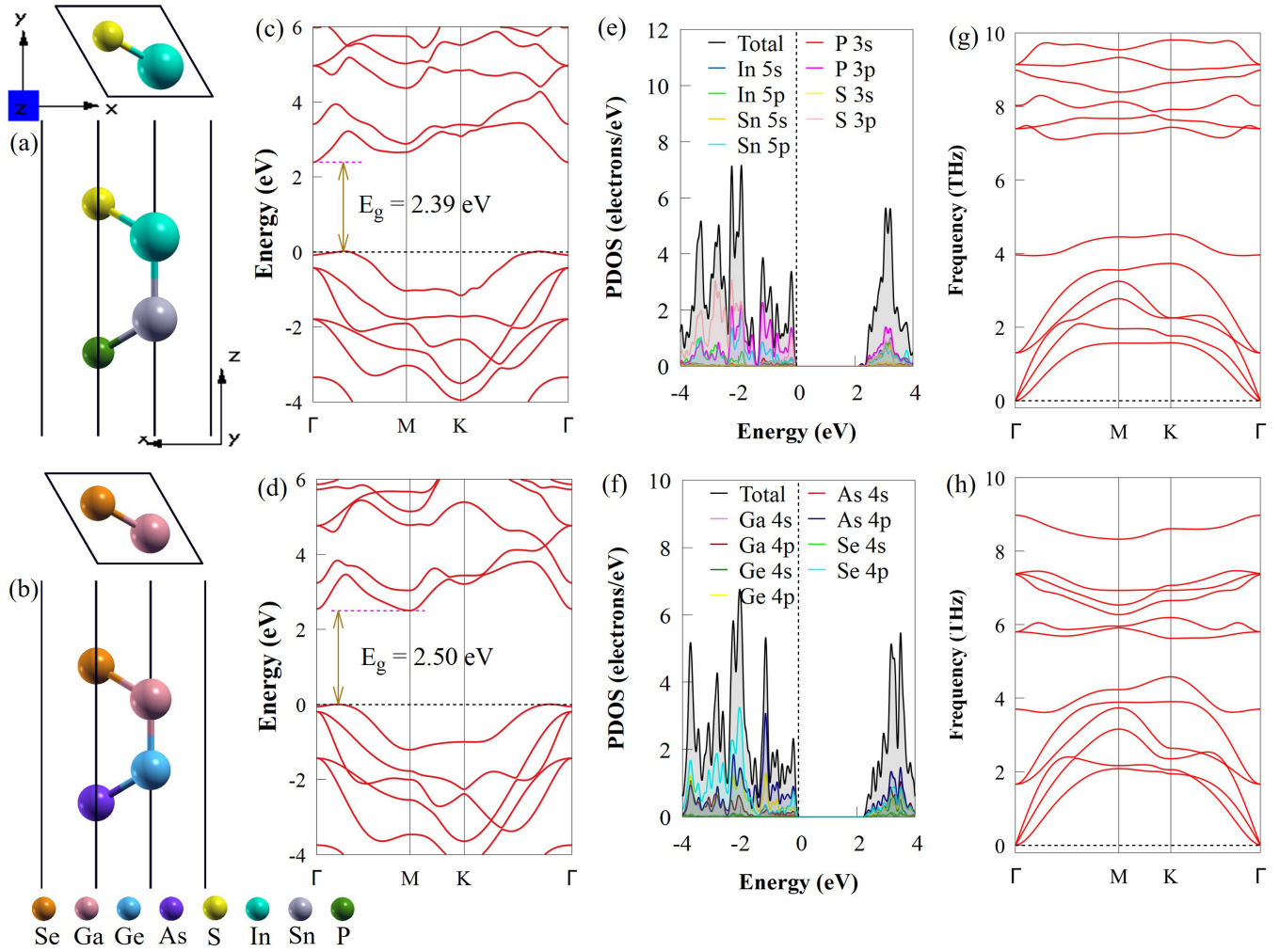


FIG. 1. Top and side views of monolayer (a) InSnPS and (b) GaGeAsSe. Band structures and projected density of states of monolayer (c), (e) InSnPS and (d), (f) GaGeAsSe. Phonon dispersion of (g) InSnPS and (h) GaGeAsSe monolayers.

CVD process has produced vertically stacked GaSe/MoSe₂ [51] misfit bilayers with large lattice mismatches, yet with well-aligned crystallographic orientation, atomically clean interfaces, and functional p-n junction behavior. Layered heterostructures of GaS and GaSe have been prepared via layered stacking and laser-based treatments [52]. Moreover, InSe/GaSe heterostructure has been grown successfully using molecular beam epitaxy [53] and high temperature pyrolysis of organometallic precursors [54]. These efforts illustrate that complex multielement 2D layers can be fabricated via CVD with precise layer alignment and quality suitable for electronic and optoelectronic applications. Synthesis of InSnPS/GaGeAsSe is experimentally feasible, building on a strong foundation from GaSe/GaS, GaSe/MoSe₂ and InS/InSe heterostructures. Based on the relative locations of atoms in InSnPS and GaGeAsSe monolayers, we examine four stacking configurations of the InSnPS/GaGeAsSe heterostructure, as shown in Fig. 2. These stacking patterns are A1, A2, A3, and A4.

The E_b value is calculated to be in a range of -15.345 to -15.761 meV/Å², indicating an exothermic process and stable InSnPS/GaGeAsSe adhesion. The results

show that the E_b is much lower than that of other vdWHs, such as PtSe₂/GaN (-5.99 meV/Å²) [55], CdS/SiH (-9.75 meV/Å²) [56], Zr₂CO₂/MoS₂ (-11.13 meV/Å²) [57], and GaN/BAs (-5.22 meV/Å²) [58], indicating that InSnPS/GaGeAsSe heterostructure possess higher stability. We found that the interlayer spacing ranges from 3.674 to 3.737 Å, which aligns with vdW interaction between the adjacent layers of a 2D heterostructure [59]. The A4 stacking configuration exhibits the lowest binding energy among the four studied patterns, indicating the most stable. Consequently, this structure is used to model the InSnPS/GaGeAsSe heterostructure in all further calculations. The cohesive energies of -4.269 eV/atom for InSnPS and -4.297 eV/atom for GaGeAsSe indicate that both monolayers are energetically stable and potentially suitable for experimental synthesis [60].

Ab Initio Molecular Dynamics (AIMD) and phonon band structure simulations are used to test the thermal and dynamic stabilities of the A4 stacking configuration [61]. After determining its structure, it is better to confirm the stability of a novel material, including its mechanical, thermodynamic, and dynamic stability. Nanostructured anode materials must withstand a broad range of operating temperatures without

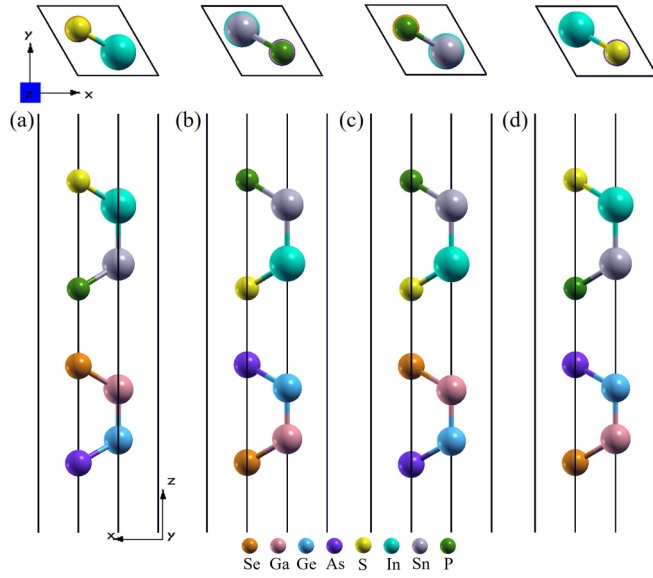


FIG. 2. Top and side views of optimized stacking configurations of InSnPS/GaGeAsSe heterostructure: stacking (a) A1, (b) A2, (c) A3, and (d) A4. Atomic species are indicated in the diagram.

suffering from heat-induced structural degradation [62]. The main element influencing how a battery operates is its structural stability. AIMD simulations on a 3×3 supercell

(72 atoms) in the A4 configuration are evaluated at 300 K to investigate the thermal stability of the InSnPS/GaGeAsSe heterostructure. Additionally, variations in the total energy are calculated using AIMD simulations. The potential energy and temperature fluctuation are plotted in Fig. 3(a). The thermal stability is demonstrated by the small energy variations and temperature fluctuation around 300 K, even for 15 ps. Following the final simulation, the structure has no broken bonds and structural reconstruction. This shows that the A4 stacking configuration is thermally stable at 300 K and confirms the viability of experimental synthesis of InSnPS/GaGeAsSe.

We used the phonon dispersion curve of InSnPS and GaGeAsSe monolayers to investigate their dynamic stability. The absence of negative/imaginary phonon modes across the Brillouin zone indicates the dynamic stability of the heterostructure and our projected 2D monolayers [Figs. 1(g) and 1(h) for the monolayers and Fig. 3(b) for the heterostructures]. Because the covalent and ionic bonds between the in-layer atoms are stronger than the vdWs forces between the layers, the majority of phonon branches in heterostructures are duplicates of the individual monolayer with minor modifications [63].

The elastic constants of $C_{11} = C_{22} = 28.99$ N/m, $C_{12} = 2.09$ N/m, and $C_{66} = 1.59$ N/m for InSnPS/GaGeAsSe meet the Born-Huang criteria ($C_{11}C_{22} - C_{12}^2 > 0$, $C_{66} > 0$) [64], proving that the InSnPS/GaGeAsSe is mechanically stable (Table II). The Young's modulus of InSnPS/GaGeAsSe heterostructure is 28.19 N/m, which is higher than that of

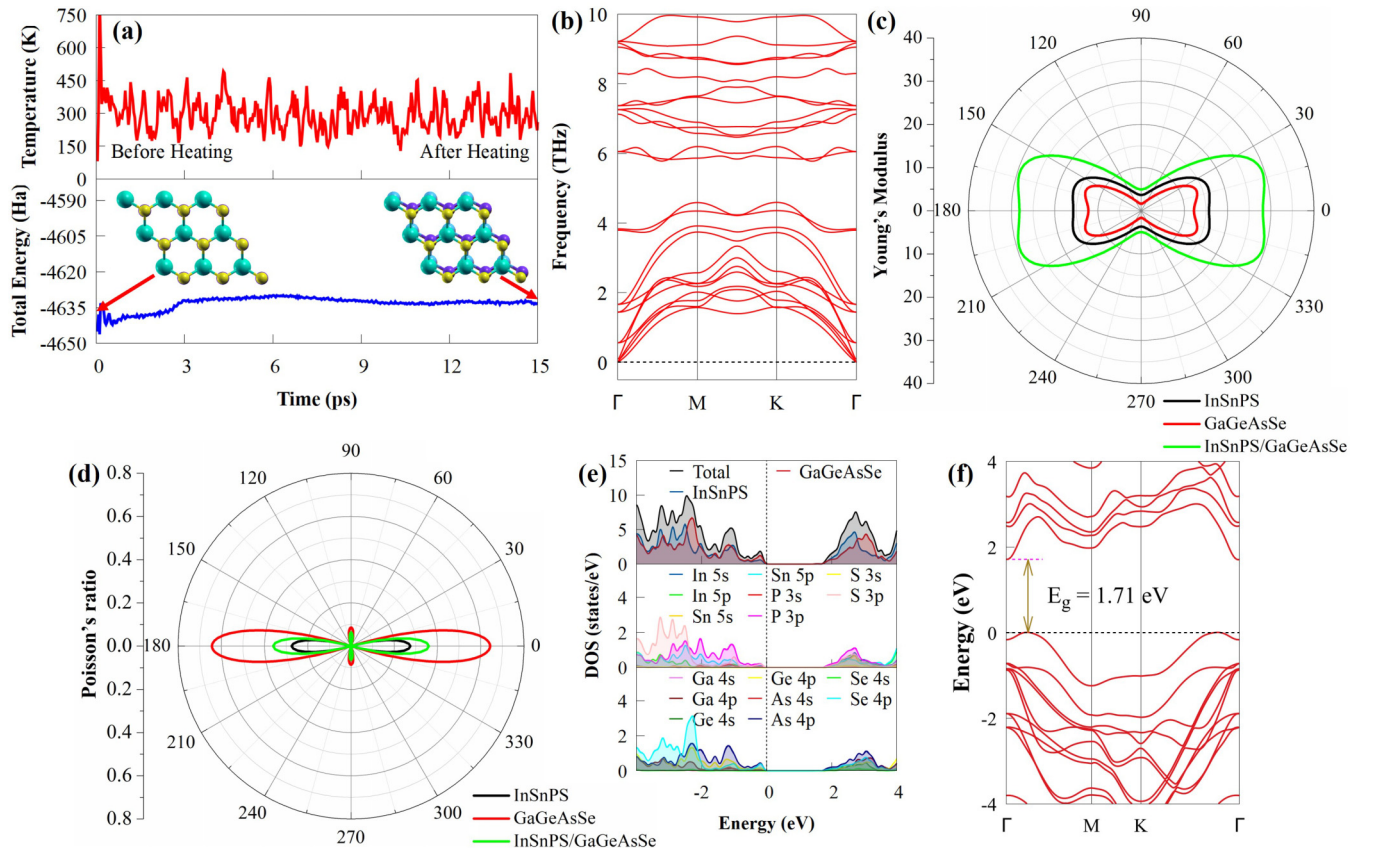


FIG. 3. (a) Fluctuations of total energy and temperature during AIMD simulation at 300 K for 15 ps, (b) Phonon dispersion spectrum, (c) angle dependent Young's modulus, (d) angle dependent Poisson's ratio, (e) Band structure, and (f) DOS for InSnPS/GaGeAsSe heterostructure as a function of the angle.

TABLE II. Calculated elastic constants $C_{11} = C_{22}$, C_{12} , C_{66} , Young's modulus (Y) and Poisson's ratio (ν) of monolayers and heterojunctions in N/m.

Material	$C_{11} = C_{22}$	C_{12}	C_{66}	Y	ν
InSnPS	16.06	1.12	0.59	15.74	0.291
GaGeAsSe	12.93	1.12	0.72	12.27	0.588
InSnPS/GaGeAsSe	28.99	2.09	1.59	28.19	0.381

InSnPS (15.74 N/m) and GaGeAsSe (12.27 N/m), proving that the InSnPS/GaGeAsSe heterostructure has good mechanical properties. The in-plane polar diagrams of the Poisson's ratio and Young's modulus are shown in Figs. 3(c) and 3(d) along various θ directions indicate that the pristine monolayers and corresponding heterostructure are anisotropic. The Young's modulus of heterostructures has consistently increased significantly compared to single components since they are thicker than the comparable monolayers [65].

The band structure for the A4 stacking configuration of the InSnPS/GaGeAsSe heterostructure is presented in Fig. 3(e). After stacking, the band structure of InSnPS/GaGeAsSe is the superposition of isolated InSnPS [Fig. 1(c)] and GaGeAsSe [Fig. 1(d)], confirming the vdWH formation. The InSnPS/GaGeAsSe has an indirect band gap (1.71 eV) semiconductor character. The VBM is situated between Γ and M symmetry points, while the CBM is situated at the Γ high-symmetry points. The electron orbitals of GaGeAsSe mainly

contribute to the orbitals near the CBM and VBM and are primarily dominated by As 4p and Se 4p orbitals [Fig. 3(f)].

As depicted in Fig. 4(a), the blue (red) isosurface denotes the depletion (accumulation) of electrons. Figure 4(b) shows the redistribution of charge, with the positive values signifying the accumulation of electrons and the negative values indicating their depletion. The electron depletion in GaGeAsSe and the electron accumulation in the P atom of InSnPS demonstrate that electrons typically transfer from the GaGeAsSe interface to InSnPS. According to the Bader charge analysis, only 0.2403 $|e|$ of charges are transferred from the GaGeAsSe interface to InSnPS. The small charge transfer highlights the weak vdW interactions in InSnPS/GaGeAsSe. Furthermore, Fig. 4(c) shows a potential drop (ΔV) of 2.91 eV between InSnPS and GaGeAsSe monolayers, demonstrating the presence of built-in electric field and its crucial role in photoexcited electron-hole separation [66].

To further analyze the local chemical bonding environments, we have computed and analyzed the electron distribution using the electron localization function (ELF) contour plot with values between 0 (blue) and 1 (red) [67]. Herein, very low charge density, gas-like pair probability and perfect localization of electrons are denoted by 0, 0.5, and 1, respectively. The calculated cleaved ELF is presented in Fig. 4(d). The value of the ELF bond point of As-P in the InSnPS/GaGeAsSe contacts is zero, indicating weak atomic orbital overlap and interfacial interactions, which further confirm the formation of vdW forces.

Work function (Φ) analysis helps gain an understanding of the conductivity of a material and can be computed using the

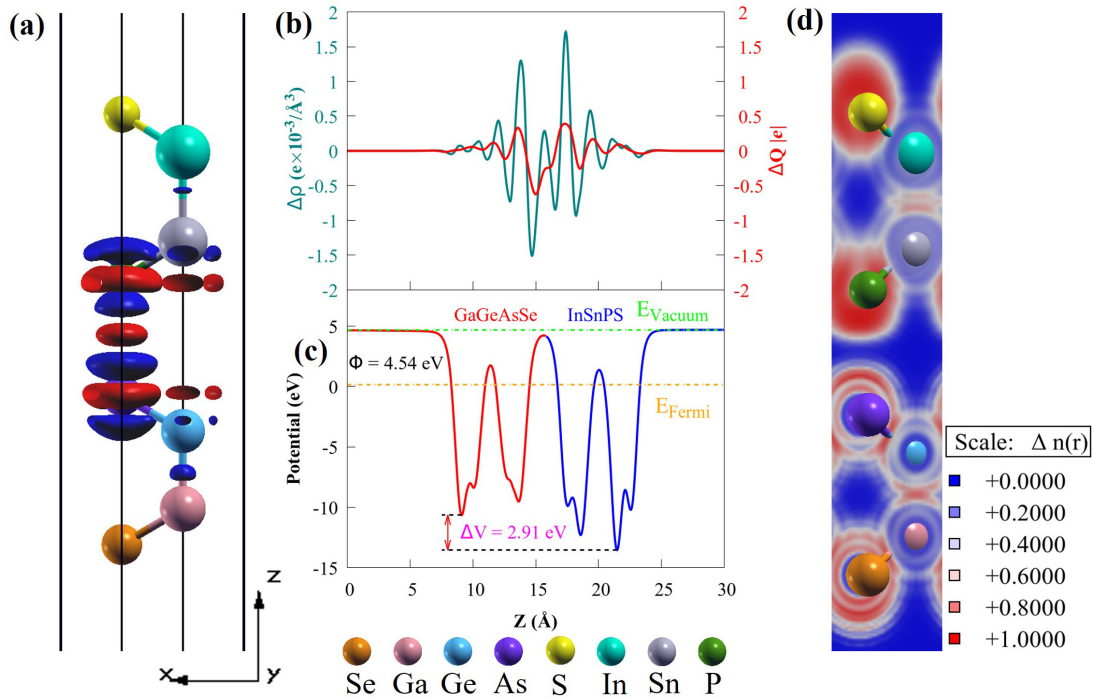


FIG. 4. (a) Side view of the 3D charge density difference of InSnPS/GaGeAsSe heterostructure with the Z-axis indicated, corresponding to the out-of-plane direction perpendicular to the layers. The isovalue is set to $8.766 \times 10^{-5} e/\text{\AA}^3$. (b) Plane-averaged charge density difference [$\Delta\rho(z)$] and (c) electrostatic potential of InSnPS/GaGeAsSe heterostructure along the Z-axis, where $\Delta V = 2.91$ eV denotes the built-in potential difference, and $\Phi = 4.54$ eV is the calculated work function. (d) The isosurface for 3D electron localization function map of InSnPS/GaGeAsSe heterostructure.

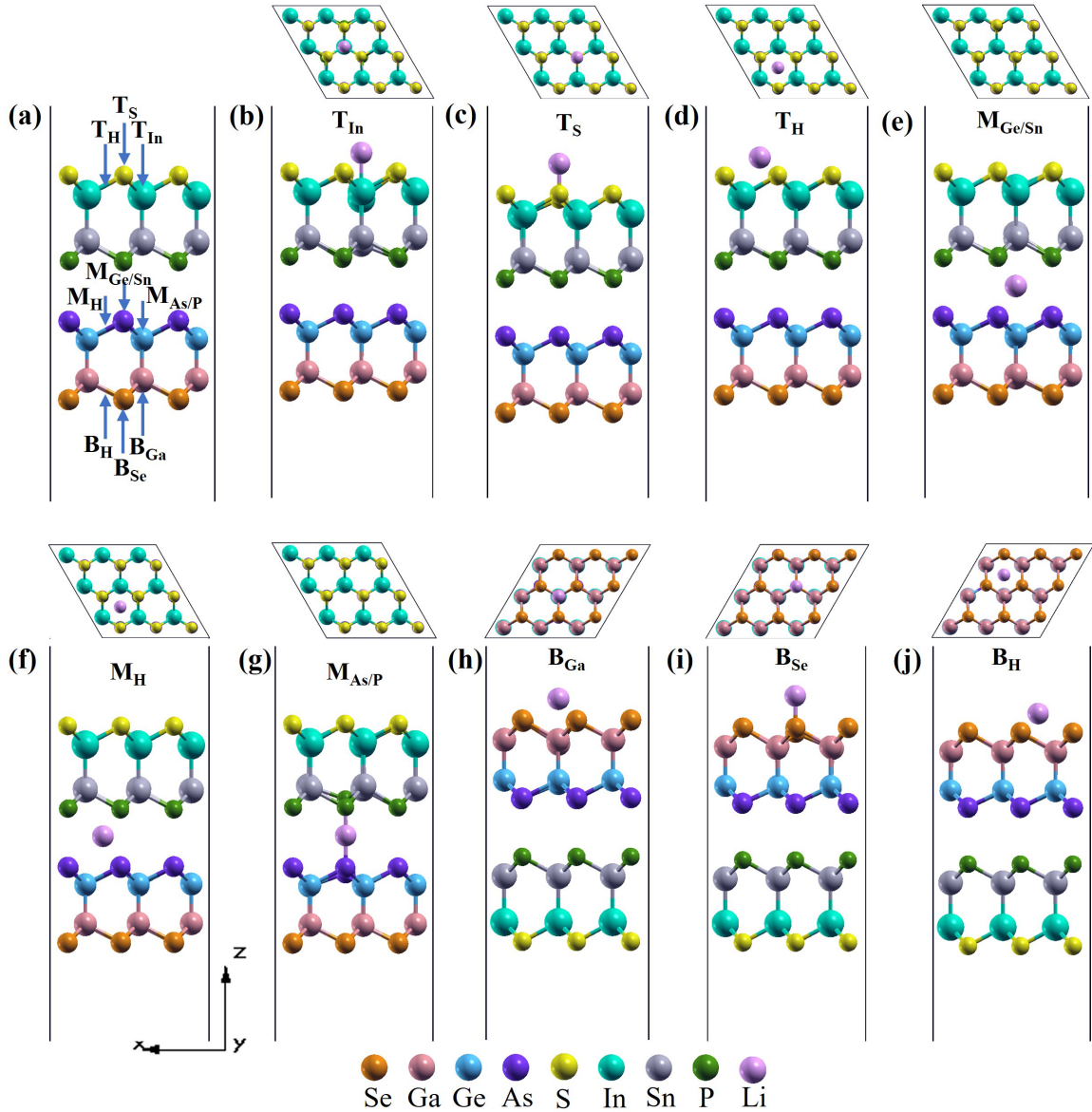


FIG. 5. (a) Possible adsorption sites of a single Li atom on InSnPS/GaGeAsSe heterostructure. Various Li intercalation configurations at the (b) T_{In} , (c) T_S , (d) T_H , (e) $M_{Ge/Sn}$, (f) M_H , (g) $M_{As/P}$, (h) B_{Ga} , (i) B_{Se} , and (j) B_H sites of InSnPS/GaGeAsSe heterostructure.

following expression [68]:

$$\Phi = E_{\text{vacuum}} - E_F \quad (10)$$

Here, E_{vacuum} and E_F refer to the vacuum and Fermi levels, respectively. The work functions of the InSnPS and GaGeAsSe monolayers are 5.94 and 5.10 eV, respectively. The model structure of InSnPS and GaGeAsSe reduces the work function value, which is 4.54 eV. InSnPS monolayer has a larger work function and lower Fermi level than the GaGeAsSe monolayer. Electrons are transferred from GaGeAsSe to InSnPS at the interface until their Fermi levels are the same, consistent with the heterostructure's charge transfer property. This confirms the reliability of our present calculations.

Assessing the adsorption behavior of metal ions in InSnPS/GaGeAsSe heterostructure requires identifying the ideal adsorption sites at/on the interface/surface [69].

We chose nine adsorption sites to test Li adatoms on the top of InSnPS (T_{In} , T_S and T_H), the bottom of GaGeAsSe (B_{Ga} , B_{Se} and B_H), and embedded at the interface of InSnPS/Li_{0.11}/GaGeAsSe ($M_{Ge/Sn}$, $M_{As/P}$ and M_H). In Fig. 5(a), the adsorption structure of a single Li atom with a potential adsorption site is displayed.

The adsorption processes for all nine adsorption sites are exothermic and energetically favorable, as indicated by the negative adsorption energy, suggesting their suitability for adatom adsorption [70]. The $M_{As/P}$ site is the most stable for single Li adsorption, with the vdWH interface enhancing adsorption strength as Li ions prefer these interfacial sites over surface locations [26].

The analysis of the isosurface along the z -axis helps visualize charge density differences, revealing that Li transfers

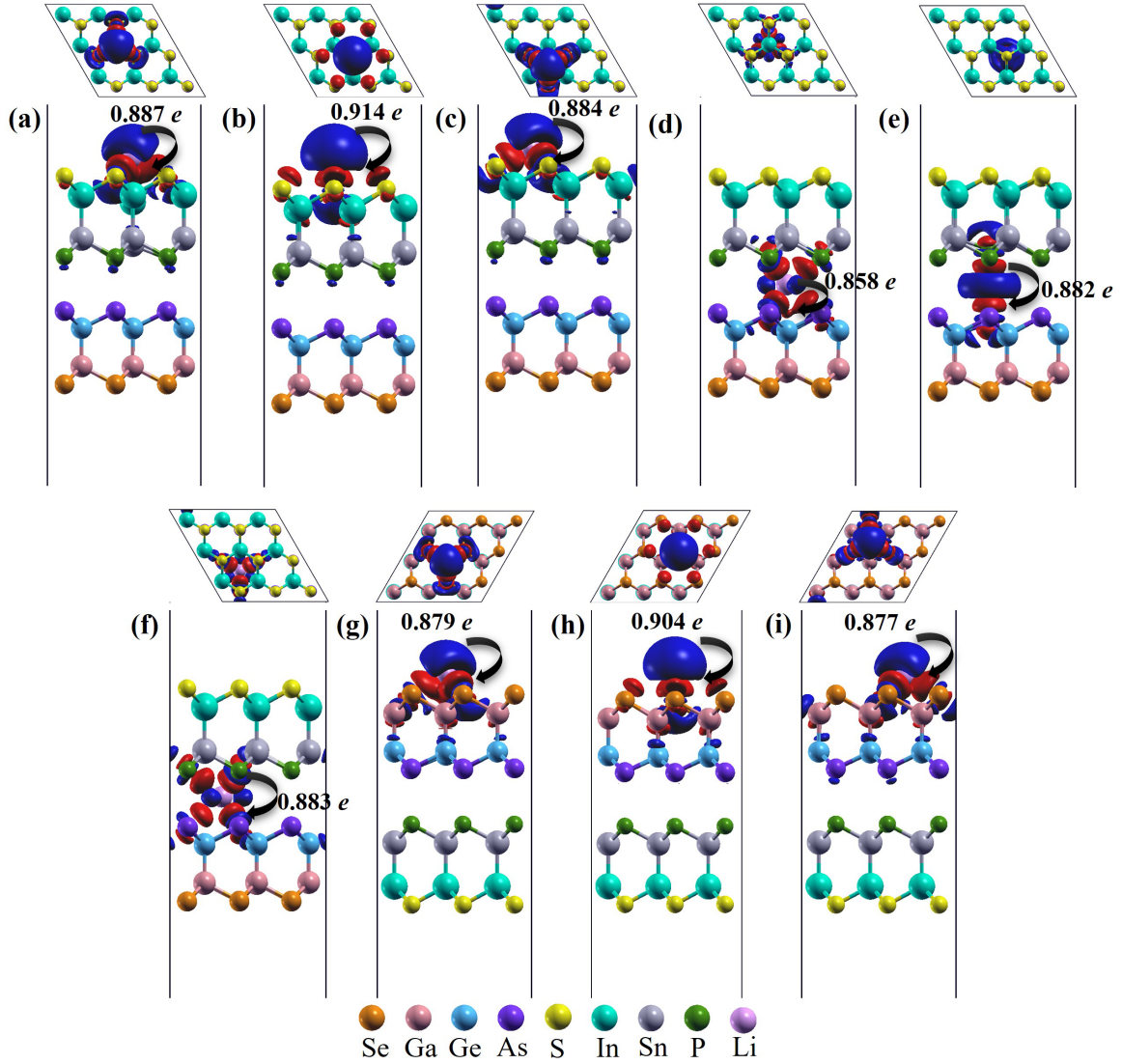


FIG. 6. Charge density difference plots for various Li intercalation configurations with large charge transfers from a single Li atom to the (a) T_{In} , (b) T_S , (c) T_H , (d) $M_{Ge/Sn}$, (e) $M_{As/P}$, (f) M_H , (g) B_{Ga} , (h) B_{Se} , and (i) B_H sites of InSnPS/GaGeAsSe heterostructure.

charge to the InSnPS/GaGeAsSe heterostructure, forming Li^+ ions (Fig. 6). Between 0.858 and 0.914 electrons are donated from Li to neighboring atoms (Table III), demonstrating

strong bonding and resulting in charge depletion around Li but accumulation near the heterostructure. The large electronegativity difference between Li and the surrounding atoms drives

TABLE III. The adsorption energy (E_{ads} , eV per Li), charge transfer (Δq , $|e|$), interface distance after adsorption (d , Å), and the distance ($Z_{\perp}$, Å) of Li from the closest atoms at the adsorption sites of the InSnPS/GaGeAsSe heterostructure.

System	Site	E_{ads} (eV)	Δq_{InSnPS} ($ e $)	$\Delta q_{GaGeAsSe}$ ($ e $)	Δq_{Li} ($ e $)	d (Å)	$Z_{\perp}$ (Å)
$Li_{0.11}/InSnPS$	T_{In}	-2.410	-0.029	-0.856	0.887	3.562	1.172
	T_S	-1.728	-0.046	-0.865	0.914	3.570	1.904
	T_H	-2.338	-0.029	-0.853	0.884	3.548	1.105
InSnPS/ $Li_{0.11}$ /GaGeAsSe	$M_{Ge/Sn}$	-2.806	-0.380	-0.501	0.883	3.593	1.723
	$M_{As/P}$	-2.856	-0.377	-0.478	0.858	3.799	1.771
	M_H	-2.709	-0.358	-0.522	0.882	3.662	1.728
	B_{Ga}	-2.301	-0.712	-0.164	0.879	3.641	1.292
GaGeAsSe/ $Li_{0.11}$	B_{Se}	-1.745	-0.720	-0.182	0.904	3.587	2.017
	B_H	-2.305	-0.708	-0.167	0.877	3.601	1.187

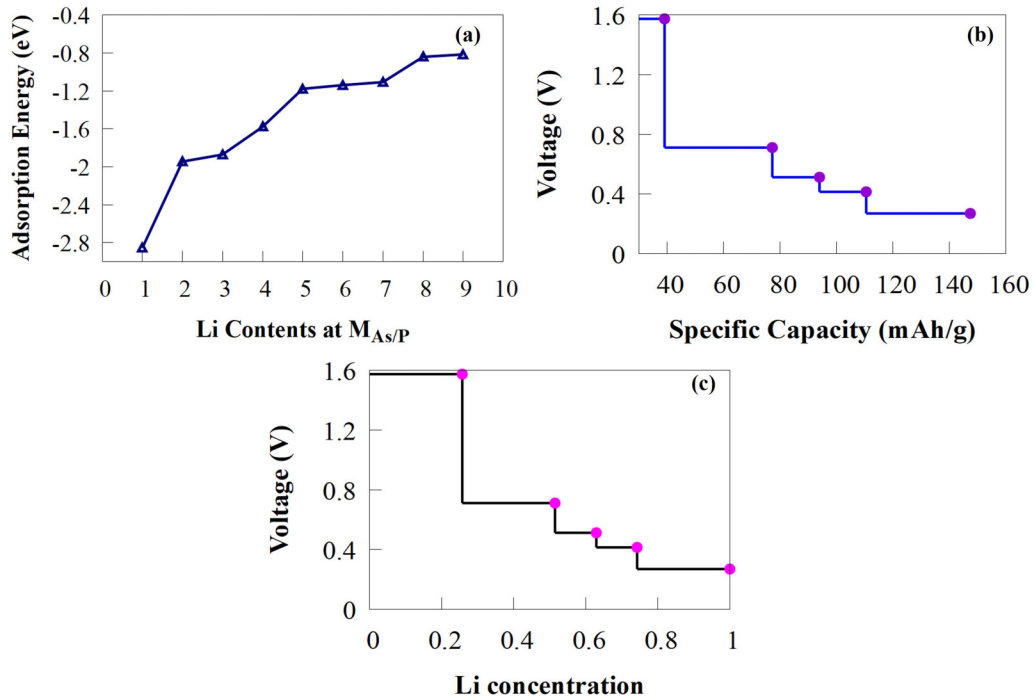


FIG. 7. (a) The adsorption energies of the InSnPS/GaGeAsSe containing nine Li adatoms on the $M_{As/P}$ site against Li ion concentration. Theoretical voltage profile for InSnPS/GaGeAsSe heterostructure against (b) specific capacity and (c) the Li ion concentration.

this significant charge transfer, averaging ~ 0.885 electrons per Li atom.

The theoretical capacity of lithium atoms, a key performance metric for anode materials in LIBs, depends on the number of Li atoms adsorbed on the InSnPS/GaGeAsSe heterostructure, influencing both specific capacity and average OCV. By analyzing maximum Li adsorption concentrations, the OCV and capacity can be determined, with initial Li adatoms preferentially occupying the most favorable interlayer sites before expanding to other locations as their numbers increase. As more Li adatoms are added, they favor interfacial sites, demonstrating the heterostructure's adsorption behavior and its implications for battery performance. As the number of adatoms increases, Li adatoms attached to different sites again favor the interface site [Fig. 7(a)].

As the number of Li concentrations increases, the average E_{ads} increases from -2.856 eV per Li atom ($n = 1$) to -0.816 eV per Li atom ($n = 9$). This results from the significant Li–Li repulsion at comparatively high Li-ion concentrations and the weak electrostatic contact between the Li ions and the InSnPS/GaGeAsSe heterostructure. The fact that the adsorption energy is still negative at the highest concentration indicates that the Li ions can fill all the sites. Lithium atoms are stacked layer-by-layer on both sides and the interface of InSnPS/GaGeAsSe heterostructure to assess the maximum theoretical capacity until all the surface-exposed sites (layers 1–5) are occupied.

The maximum storage capacity of InSnPS/GaGeAsSe is calculated to be $483.38 \text{ mAh g}^{-1}$, which is higher than that of $\text{V}_2\text{CO}_2/\text{graphene}$ (234 mAh g^{-1}) [71], MoS_2/WS_2 (137 mAh g^{-1}) [72], Si/C (278.4 mAh g^{-1}) [73], $\text{MoS}_2/\text{Ti}_2\text{CO}_2$ (447 mAh g^{-1}) [24], antimonene/graphene (369 mAh g^{-1}) [74], and MoSSe/graphene (390 mAh g^{-1})

[75]. Figure 7(b) illustrates the stable yet fluctuating relationship between the OCV and the specific capacity during this process. Additionally, Fig. 7(c) demonstrates how the OCV declines as the number of Li layers increases, with values ranging from 1.572 to 0.269 V. Notably, no Li dendrites form during lithiation, ensuring small but positive voltages throughout the adsorption process. This characteristic enhances the energy density and operating voltage of LIBs. The conclusion that no Li dendrites form during lithiation arises from the observed adsorption and diffusion behavior of Li atoms in the studied system. The Li ions are uniformly adsorbed at energetically favorable sites and diffuse smoothly through the host structure without clustering or forming filament-like growths. The calculated adsorption energies and migration pathways indicate homogeneous lithiation rather than localized accumulation, which typically drives dendrite nucleation. Additionally, the absence of structural distortion or sharp anisotropy in Li distribution across the layers supports the inference that dendritic growth is suppressed in this material. The InSnPS/GaGeAsSe heterostructure exhibits an outstanding average OCV of 0.69 V, which is within the ideal 0–1 V range for LIB anodes and lower than that of $\text{TiS}_2/\text{Ti}_2\text{CS}_2$ (1.663 V), [76] $\text{MoS}_2/\text{Ti}_2\text{CS}_2$ (0.79 V) [26], and $\text{MoS}_2/\text{Ti}_2\text{CO}_2$ (0.82 V) [24] heterostructures.

To assess the maximum theoretical capacity, lithium atoms are stacked layer-by-layer on both sides and interface of InSnPS/GaGeAsSe heterostructure until all the surface-exposed sites (layers 1–5) are occupied, see Figs. 8(a)–8(e).

On a 3×3 supercell of the InSnPS/GaGeAsSe heterostructure, Li atoms can be adsorbed in an ordered manner. The fully lithiated InSnPS/GaGeAsSe is shown in Fig. 8(e). In the initial adsorption stage, Li atoms occupy all interface sites on the surface (first Li adsorption layer) with an E_{ads} of

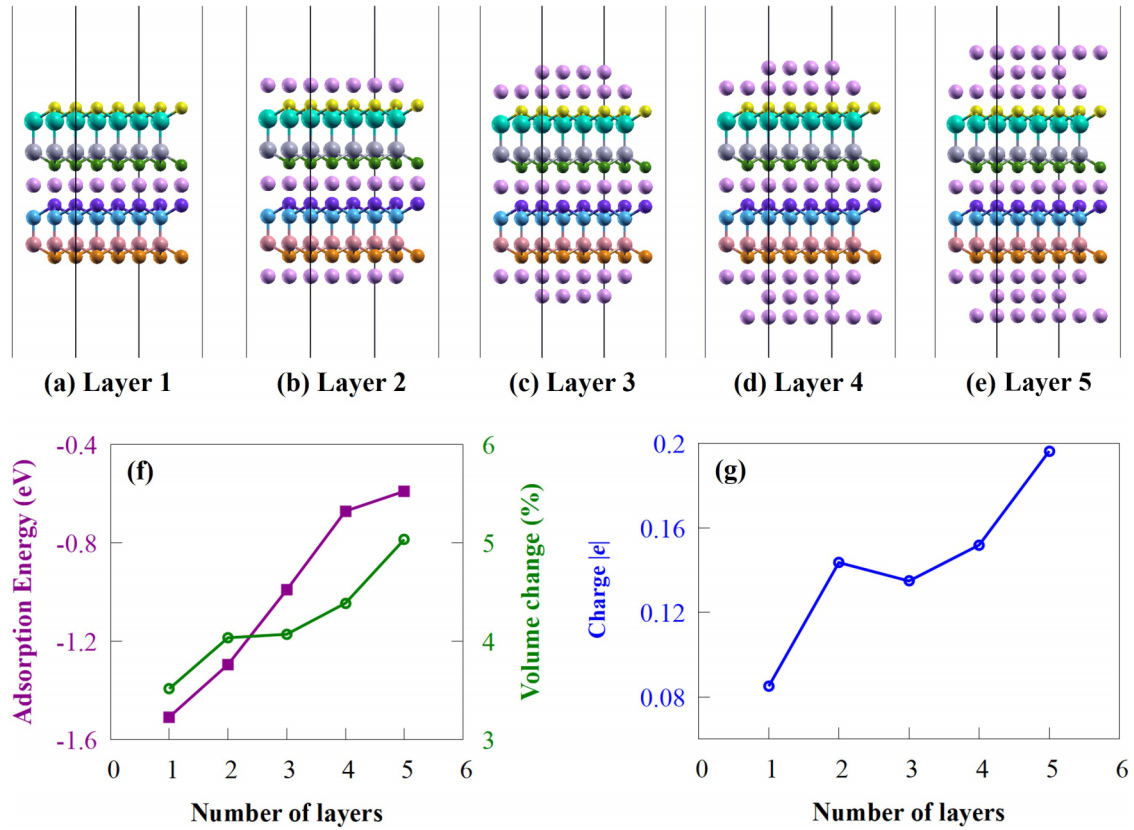


FIG. 8. (a)–(e) Top and side views of layer n of Li ions adsorbed on InSnPS/GaGeAsSe ($n = 1, 2, 3, 4$, and 5). (f) Adsorption energies (purple line with points) and volume change (green line with points) and (g) the charge transfer of InSnPS/GaGeAsSe against Li-ion concentration.

–1.094 eV. Successive adsorption layers were considered by adding additional planes of Li atoms above the first, yielding E_{ads} values of –0.786 eV for the second layer. With increasing number of Li adsorption layers, the E_{ads} gradually increases from –1.094 eV per Li atom (first layer) to –0.546 per Li atom (fifth layer), as shown in Fig. 8(f). The progressive decrease in binding strength with additional Li adsorption layers illustrates the lithiation process and surface saturation behavior. All the structures showed negative adsorption energy values and were fully relaxed, indicating that the Li atoms can be stable adsorbed on InSnPS/GaGeAsSe surface. An InSnPS/GaGeAsSe structure with Li atoms between the layers is used to estimate the distance change of the InSnPS/GaGeAsSe after Li intercalation. This allows for the determination of the volume expansion of InSnPS/GaGeAsSe during the lithiation process, as shown in Fig. 8(f). The volume expansion values at layers 1, 2, 3, 4, and 5 are 3.52, 4.04, 4.07, 4.39, and 5.04%, respectively. Li intercalation induces a volume expansion of 5.04%, with the lattice parameter increasing from 3.674 Å in the pristine InSnPS/GaGeAsSe structure to 3.859 Å after complete lithiation. This expansion is significantly lower than the 10% predicted for graphite [77]. Moreover, charge transfer increases with an increase in the number of layers for Li loading [Fig. 8(g)]. This proves that the InSnPS/GaGeAsSe can adsorb five-layer Li atom at least.

The mobility of Li ions on a material's surface is a key factor in evaluating its suitability as an electrode, with lower diffusion barriers enhancing charge-discharge rates.

Although our study focuses on the activation barriers for Li-ion migration, these barriers can be further employed, together with an adequate energy potential, to estimate the corresponding diffusion coefficient variations with Li content, as demonstrated by Gavilán-Arriazu *et al.* [78], thereby providing a more quantitative link to Li-ion transport properties and battery performance. To study Li-ion mobility in the InSnPS/GaGeAsSe heterostructure, we analyzed the diffusion of a single Li atom at the top, interlayer, and bottom surfaces [Figs. 9(a)–9(c)].

Additionally, as seen in Figs. 9(d) and 9(e), we examined the diffusion barrier energies and computed the lowest energy pathway of Li atoms diffusing on the different surfaces and interfaces of the heterostructure.

Comparably, the B_H site is the most stable one at the GaGeAsSe surface; as a result, the migration barrier from the B_H site to another is computed, and the barrier energy is 0.23 eV due to the small potential energy difference between B_H sites. The most stable site for lithiation at the interface is the $M_{\text{As/P}}$, and the barrier energy for diffusing Li between $M_{\text{As/P}}$ sites is 1.62 eV. Li migration on GaGeAsSe is thought to be an energetically favorable path. When considering the adsorption energy (–2.856 eV), Li intercalation is stronger at the interface. Similarly, the GaGeAsSe and InSnPS layer interface has the largest diffusing barrier for Li ions, the same as reported in other heterostructures [65]. InSnPS/GaGeAsSe has fast charge/discharge rates that could be anode materials for LIB when compared to the 0.45 eV for MoS_2/VS_2 [79],

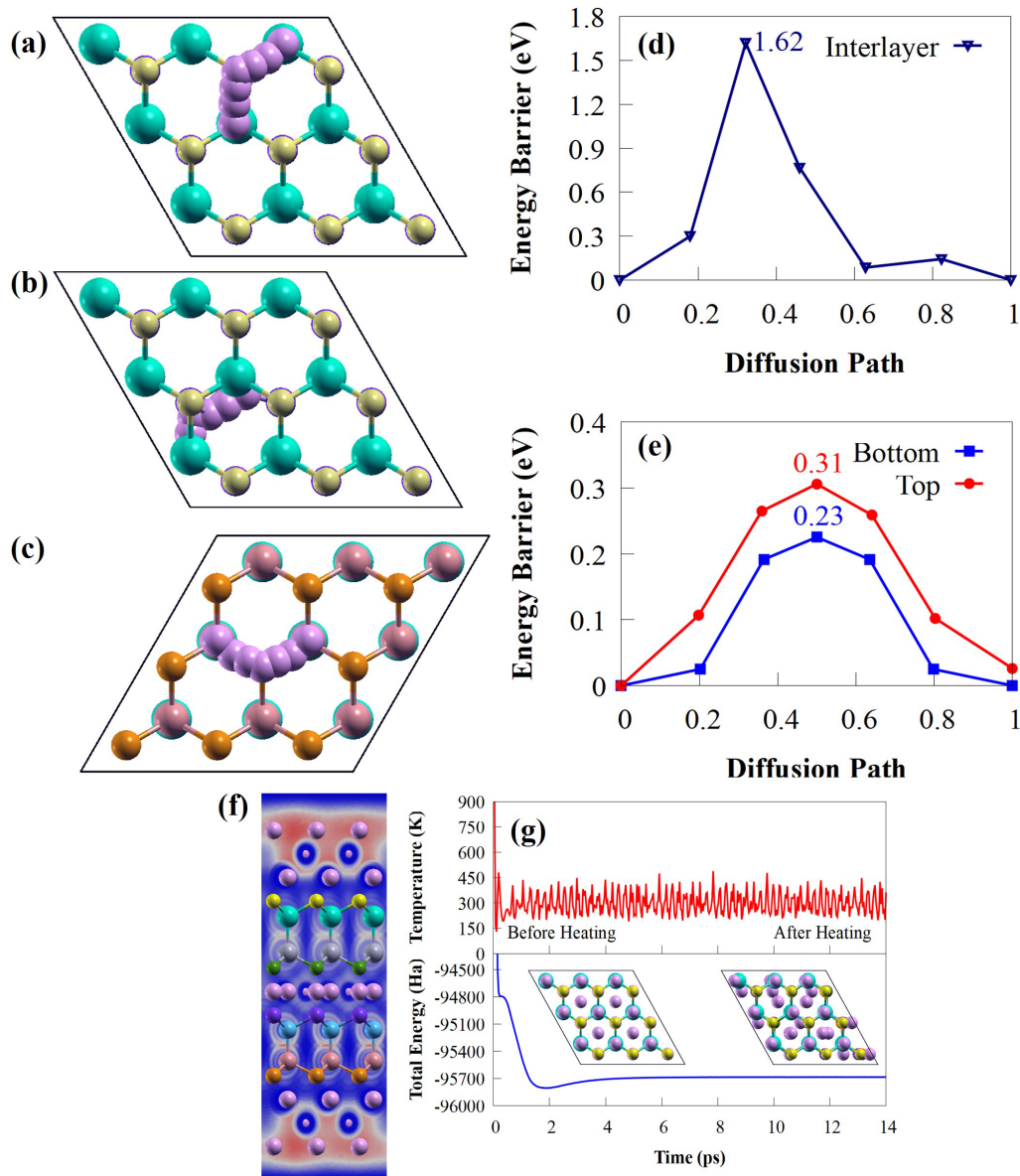


FIG. 9. (a-c) Diffusion pathways of a single Li^+ ion at the top, interface, and bottom of the InSnPS/GaGeAsSe heterostructure. Diffusion barrier of a single Li^+ ion (d) at the interface and (e) on the surface of the InSnPS/GaGeAsSe heterostructure. The inset values indicate the corresponding diffusion barrier energies. (f) ELF map of fully lithiated InSnPS/GaGeAsSe heterostructure. (g) Total potential energy and temperature fluctuations during the AIMD simulation of fully lithiated InSnPS/GaGeAsSe heterostructure after 14 ps simulation time.

0.41 eV for $\text{VS}_2/\text{Ti}_2\text{CO}_2$ [25], 0.60 eV for $\text{V}_2\text{CO}_2/\text{graphene}$ [80], and 0.57 eV for $\text{MoS}_2/\text{Ti}_2\text{CO}_2$ [24]. A small activation energy promotes rapid ion migration, benefiting charge and discharge processes. However, to suppress leakage current, it is equally important to retain a large electronic band gap together with structural robustness. Achieving this balance is key to combining high ionic conductivity with reliable performance and safety in real device applications.

To examine the bonding properties, the ELF maps of fully lithiated InSnPS/GaGeAsSe are plotted, as seen in Fig. 9(f). It can be observed that the InSnPS/GaGeAsSe has more localized electrons around the Li-top/bottom-most atoms, indicating a stronger interaction. The enormous number of electrons that are localized between the Li adatom layer and the InSnPS/GaGeAsSe substrate reveals that ionic

bonds exist between them. The stability of the fully lithiated InSnPS/GaGeAsSe heterostructure is then confirmed using AIMD simulations. After operating at 300 K for 14 ps, the 35 Li atoms are still adsorbed on InSnPS/GaGeAsSe heterostructure. The lithiated InSnPS/GaGeAsSe remains intact regardless of the minor structural changes displayed in Fig. 9(g). Moreover, the total energy and temperature fluctuate around a fixed value, indicating its thermal stability [Fig. 9(g)]. Therefore, it can be concluded that fully lithiated InSnPS/GaGeAsSe maintain structural stability at room temperature and can be considered excellent anode materials for LIBs.

To further understand the intercalation of a Li atom with a host material, we showed in Fig. 10 the PDOS of InSnPS/GaGeAsSe heterostructure with a Li ion adsorbed. The

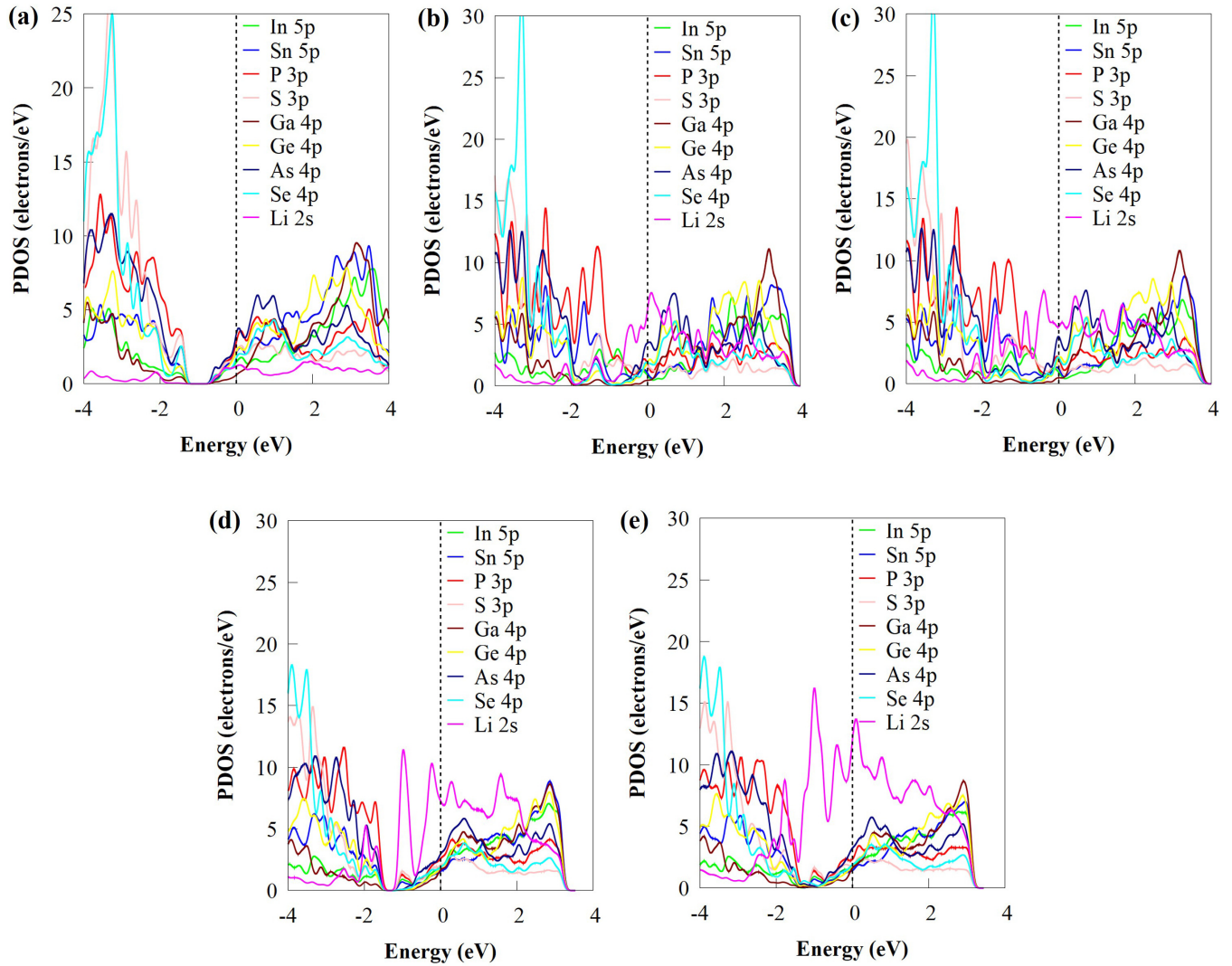


FIG. 10. Density of states plots for Layer (a) 1, (b) 2, (c) 3, (d) 4, and (e) 5 upon Li intercalation in the InSnPS/GaGeAsSe heterostructure.

electrical conductivity of the anode is another crucial factor affecting the batteries' performance and rate capability. When the Li atom is fully adsorbed, it contributes significantly to the hybridization between the InSnPS/GaGeAsSe and Li [Fig. 10(e)], strengthening the heterostructures' strong interaction with Li. After a metal ion is fully adsorbed, the PDOS of the InSnPS/GaGeAsSe heterostructure essentially rises, facilitating electron transmission and satisfying the high conductivity requirements of batteries.

The metallicity of the Li-incorporated InSnPS/GaGeAsSe heterostructure demonstrates the charge transfer from Li to the InSnPS/GaGeAsSe. This characteristic guarantees its significance in the application as a LIB with significantly improved conductivity, enabling a quick charging and discharging process.

IV. CONCLUSIONS

Using a DFT study, we designed the InSnPS/GaGeAsSe heterostructure and systematically examined its prospect as an anode material for rechargeable LIBs. Mechani-

cal and phonon dispersion simulations verified the InSnPS/GaGeAsSe heterostructure's strong mechanical and dynamic stabilities. According to our calculations, the InSnPS and GaGeAsSe monolayers are stable because of their negative formation energy. The specific capacity of the fully lithiated InSnPS/GaGeAsSe heterostructure was $483.38 \text{ mAh g}^{-1}$. Compared to previous LIB heterostructures, the diffusion energy barrier at the bottom is lower, suggesting superior rate capacity. According to the DOS results, the InSnPS/GaGeAsSe heterostructure has superior electronic conductivities, making it suitable for Li-ion energy storage. In addition, the low OCV, high conductivity and good cycle stability make the InSnPS/GaGeAsSe heterostructure an excellent anode material for LIBs.

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The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

DATA AVAILABILITY

The data that support the findings of this article are not publicly available. The data are available from the authors upon reasonable request.

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