M_2X Monolayers as Anode Materials for Li Ion BatteriesAbdus Samad[✉] and Udo Schwingenschlöggl^{*}*Physical Science and Engineering Division, King Abdullah University of Science and Technology (KAUST), Thuwal 23955-6900, Saudi Arabia*

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Electrochemically efficient electrode materials are required for clean energy storage in Li ion batteries. We predict two-dimensional hexagonal metal nitrides, borides, and phosphides (Sc_2B , Sc_2N , Y_2B , Y_2N , and Y_2P) and evaluate the feasibility of experimental realization. The materials combine excellent metallicity, as required for electrodes, with Li binding energies providing high storage capacity and a low average open-circuit voltage. In contrast to two-dimensional silicene, borophene, and SnS_2 , we observe negligible structural distortions during Li adsorption and extraction, which results in high reversibility and a long cycle life. Superionic Li diffusion enables fast charge or discharge of next-generation Li ion batteries.

DOI: [10.1103/PhysRevApplied.15.034025](https://doi.org/10.1103/PhysRevApplied.15.034025)**I. INTRODUCTION**

Rechargeable Li ion batteries (LIBs) play a crucial role as the power supply of electric vehicles and portable electronics, providing high energy density, high power rate, long cycle life, and low self-discharge [1,2]. The electrochemical performance depends on the anode, cathode, and electrolyte, which can be optimized individually to a large extent [3]. Concerning anodes, the storage capacity of conversion and alloying anodes is usually higher than that of intercalation anodes but a limited cyclability precludes commercial use [4–7]. Graphite, the conventional anode material of commercial LIBs, provides a storage capacity of 372 mAh/g due to reversible intercalation of Li ions in the layered structure [4]. Two-dimensional graphene, on the other hand, suffers from weak Li binding (low storage capacity), which can be enhanced through defects only by sacrificing the Li diffusion [8–11]. Graphene composite materials [12–14] presumably will be too expensive for usage in commercial LIBs. Other two-dimensional materials face issues of structural distortions during Li loading, weak Li binding, low metallicity, and/or slow Li diffusion [15–17]. Black arsenic [18] and MoO_2 , MoS_2 , MoSe_2 , WO_2 , WS_2 , and WSe_2 [19,20] monolayers lack electronic conductivity and/or suffer from poor cyclability [21,22].

Two-dimensional metal carbides and metal nitrides (Mo_2C , W_2C , Mo_2N , etc.) provide high stiffness, high electronic and ionic mobilities, high Li storage capacity, and superionic conductivity (which enables low charge or discharge times and high power rates) [23–29]. In the present study, we predict the related Sc_2B , Sc_2N , Y_2B , Y_2N , and Y_2P monolayers and evaluate their application

potential in LIBs. Experimental feasibility of synthesis is demonstrated in terms of cohesive energies, phonon spectra, and mechanical and thermal stabilities. The Sc_2N and Y_2N monolayers exhibit suitable Li binding energies for high Li storage capacity. Negligible structural changes during Li loading, low average open-circuit voltage, excellent metallicity, and superionic Li diffusion make them highly promising anode materials for LIBs.

II. METHODS

First-principles calculations within the framework of density-functional theory are used to predict the structural, electronic, and Li adsorption and diffusion properties [30–32], employing the projector-augmented-wave method of the Vienna *ab initio* simulation package [33]. The electron exchange-correlation interaction is modeled in the generalized gradient approximation (Perdew-Burke-Ernzerhof functional) and the van der Waals interaction is taken into account by the method of Ref. [34]. The energy cutoff of the plane-wave expansion is set to 520 eV and for structure optimization a force criterion of 0.001 eV/Å is used. Each slab model contains a vacuum layer of 15 Å thickness in the out-of-plane direction and Γ -centered $25 \times 25 \times 1$ k meshes [35] are used for Brillouin-zone integration. The PHONOPY code is used to calculate phonon spectra [36]. The elastic constants are obtained by the stress-strain approach [37]. *Ab initio* molecular dynamics simulations are conducted for $6 \times 6 \times 1$ supercells of the M_2X monolayers using a canonical ensemble and the Nosé-Hoover thermostat. Also, $3 \times 3 \times 1$ supercells to achieve low Li coverage are used for studying the Li

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adsorption and determining the saddle points and minimum energy Li diffusion pathways by the climbing-image nudged-elastic-band method [38].

III. RESULTS AND DISCUSSION

As two-dimensional metal carbides and metal nitrides realize either 1T or 2H hexagonal structures [24–27,39], we consider both of these possibilities for the Sc_2B , Sc_2N , Y_2B , Y_2N , and Y_2P monolayers. We find in each case that the 1T structure is energetically favorable and therefore we study it in the following. The obtained lattice parameters are given in Table I together with the energy differences compared with the corresponding 2H structures. We find in quasiharmonic approximation that a temperature of 300 K alters the lattice parameters by less than 0.03 Å. As an example, top and side views of the Y_2N monolayer are shown in Fig. 1(a). We determine the cohesive energy as $E_{\text{coh}} = [2E_M + E_X - E_{M_2X}]/3$, where E_M and E_X are the spin-polarized total energies of isolated M and X atoms, respectively, and E_{M_2X} is the total energy of the M_2X monolayer. The results are given in Table I. They are compared to results for the MoS_2 and SnS_2 monolayers obtained by the same methodology. For all M_2X monolayers, E_{coh} is higher than for the SnS_2 monolayer, and for the Sc_2N and Y_2N monolayers it is also higher than for the MoS_2 monolayer.

The phonon spectra in Figs. 1(b)–1(f) show three acoustic and six optical modes, demonstrating dynamical stability of the M_2X monolayers due to the absence of negative frequencies. For the Y_2P monolayer, we also find dynamical stability for the 2H structure. The obtained elastic constants (C_{11} , C_{12}), the Young's moduli (Y), and the Poisson ratios (ν) are given in Table I. The Young moduli show that the stiffness of the Sc_2N monolayer exceeds that of the MoS_2 monolayer, while the results for the other M_2X monolayers fall between those of the SnS_2 and MoS_2 monolayers. All monolayers satisfy the Born criteria of mechanical stability ($C_{11} > 0$, $C_{11} > |C_{12}|$) [40]. *Ab initio* molecular-dynamics simulations conducted for the Sc_2N and Y_2N monolayers (which provide suitable Li binding energies, see below) prove thermal stability at 300 K [see Figs. 2(a) and 2(b)], as the structures are conserved even

FIG. 1. (a) Top and side views of the Y_2N monolayer (1T structure). Phonon spectra of the M_2X monolayers: (b) Sc_2B ; (c) Sc_2N ; (d) Y_2B ; (e) Y_2N ; (f) Y_2P .

after 6 ps of simulation time. For the layered bulk phases of Sc_2N and Y_2N (Fig. S1 [41]) with $\text{P}\bar{3}m1$ symmetry, we obtain cleavage energies (energy difference between the bulk phase per layer and the isolated layer) of 0.32 and 0.24 Jm^{-2} , respectively, after optimization of the lattice parameters ($a = 3.20$ Å and $c = 5.35$ Å for Sc_2N with an interlayer distance of 2.90 Å; $a = 3.47$ Å and $c = 5.91$ Å for Y_2N with an interlayer distance of 3.25 Å). These values are significantly smaller than the cleavage energy of 2.15 Jm^{-2} of the experimentally exfoliated

TABLE I. The lattice parameter (Å), cohesive energy (eV per atom), energy difference per formula unit between the 1T and 2H structures (eV), elastic constants (N/m), Young's modulus (N/m), Poisson ratio, and Li binding energies at the H and T sites (meV).

	a	E_{coh}	ΔE	C_{11}	C_{12}	Y	ν	E_b^{H}	E_b^{T}
MoS_2	3.18	5.28	0.85	132.5	32.8	124.4	0.24	...	...
SnS_2	3.69	3.67	−0.86	82.5	29.1	72.2	0.35	...	...
Sc_2B	3.61	3.82	−0.87	83.6	26.5	75.2	0.32	−111	−111
Sc_2N	3.18	5.43	−0.57	135.7	36.6	125.8	0.27	−524	−524
Y_2B	3.84	3.79	−0.72	67.7	24.0	59.2	0.36	−30	−25
Y_2N	3.41	5.36	−1.23	117.0	34.6	106.8	0.29	−276	−275
Y_2P	3.78	4.75	−0.66	85.3	23.8	78.7	0.28	−109	−102

FIG. 2. The temperature (black) and total energy (blue) during the *ab initio* molecular-dynamics simulations of the (a) Sc_2N and (b) Y_2N monolayers at 300 K. The insets show the atomic structures after 6 ps of simulation time.

ReS_2 monolayer [42]. In contrast to many two-dimensional materials, the electronic band structures of the M_2X monolayers in Fig. 3 demonstrate excellent metallicity, which is one of the most important requirements of an anode material.

We now study the Li storage capacity, the structural distortions during Li adsorption, the average open-circuit voltage, and the Li diffusion barrier. The Li binding energy is defined as $E_b = E_{M_2X+\text{Li}} - E_{M_2X} - E_{\text{Li}}$, where $E_{M_2X+\text{Li}}$ and E_{M_2X} are the total energies of the M_2X monolayer with and without the Li atom adsorbed, respectively, and E_{Li} is the total energy of an atom in the body-centered-cubic structure of bulk Li. Thus, a negative value means that Li adsorption results in an energy gain. Among the tested adsorption sites, the hollow (H) site above the bottom M atoms of the M_2X monolayer and the top (T) site above the X atoms [compare Figs. 4(a) and 4(b)], turn out to be energetically favorable over the bridge site connecting the M and X atoms and the top site above the M atoms. According to Table I, similar E_b values are obtained for a single Li atom adsorbed at the H and T sites, which is due to the similar binding environments. As the Sc_2N and Y_2N monolayers provide the strongest bonding, we further consider them as anode materials for LIBs. Note that Li intercalation in bulk Sc_2N and Y_2N is thermodynamically unfavorable. We determine the valence-charge-density difference $\Delta\rho = \rho_{M_2X+\text{Li}} - \rho_{M_2X} - \rho_{\text{Li}}$, where $\rho_{M_2X+\text{Li}}$ and

FIG. 3. The electronic band structures (spin-orbit coupling included) of the M_2X monolayers: (a) Sc_2B ; (b) Sc_2N ; (c) Y_2B ; (d) Y_2N ; (e) Y_2P .

ρ_{M_2X} are the valence charge densities of the M_2X monolayer with and without the Li atom adsorbed, respectively, and ρ_{Li} is the valence charge density of an isolated Li atom. Figures 4(c) and 4(d) show, for the Sc_2N and Y_2N monolayers, top and side views of $\Delta\rho$ for Li adsorption at the T site, demonstrating a large charge transfer (0.87 and 0.82 electrons, respectively, according to Bader analysis).

We calculate the average Li adsorption energy (total energy difference per atom before and after loading) to estimate the Li storage capacity, considering simultaneous adsorption on both sides of the monolayer as well as the possibility of multilayer adsorption. For this purpose, we employ a $2 \times 2 \times 1$ supercell and assume that volume and entropy effects are negligible during the adsorption. The Li atoms occupy H sites in the first layer and T sites in the second layer. As eight atoms can be accommodated in each Li layer, we define the average Li adsorption energy of the n th layer as $E_{\text{av}}(n) = (E_{M_2X+8n\text{Li}} - E_{M_2X+(n-1)\text{Li}} - 8E_{\text{Li}})/8$, where $E_{M_2X+8n\text{Li}}$ and $E_{M_2X+(n-1)\text{Li}}$ are the total energies of the M_2X monolayer loaded with n and $n-1$ Li layers, respectively, and E_{Li} is the total energy of an atom in the body-centered-cubic structure of bulk Li. For the Sc_2N monolayer, we obtain for the first, second, and third Li layers (Fig. S2 [41]) values of -458 , -22 , and -8 meV, respectively. The fact that E_{av} becomes positive

FIG. 4. Top and side views of the (a) H and (b) T adsorption sites and the valence-charge-density difference induced by Li adsorption on the (c) Sc₂N and (d) Y₂N monolayers (yellow, charge accumulation; blue, charge depletion; isovalue = 10^{-3} electrons/Å³).

for the fourth Li layer shows that a maximum of three Li layers can be stored (Li₆Sc₂N). For the Y₂N monolayer, we obtain only for the first Li layer a negative value of -255 meV, implying that only one Li layer can be stored (Li₂Y₂N).

The Li storage capacity is defined as $C = xzF/M$, where x is the number of Li ions stored per formula unit, $z = 1$ is the ionic charge, $F = 26\,801$ mAh/mol is the Faraday constant, and M is the molar mass of the M_2X monolayer. We obtain 1547 mAh/g for the Sc₂N monolayer and 279 mAh/g for the Y₂N monolayer. The Li storage capacity of the Sc₂N monolayer thus clearly exceeds literature reports for the MXenes Mo₂C (526 mAh/g) [25] and Ti₃C₂ (320 mAh/g) [43], the transition-metal dichalcogenide TiS₂ (479 mAh/g) [44], and the heterostructures MoS₂/VS₂ (584 mAh/g) [15] and MoS₂ per graphene (338 mAh/g) [45]. Moreover, the average open-circuit voltage $V_{av}(n) = E_{av}(n)/e$ ranges between 0.46 and 0.02 V for the Sc₂N monolayer and amounts to 0.22 V for the Y₂N monolayer. These low values are favorable for high-voltage LIBs.

FIG. 5. The electron localization functions of the (a) Sc₂N and (b) Y₂N monolayers with one adsorbed Li layer.

While the alloying mechanism gives rise to the highest Li storage capacities, graphite (insertion mechanism [4,5,7]) is still the first choice for commercial LIBs because of its high reversibility and long cycle life. Generally, structural changes in the anode material during lithiation and delithiation reduce the reversibility and the cycle life. In the cases of the SnS₂ and TiS₂ monolayers, the presence of Li affects the Sn—S and Ti—S bonds, respectively, and leads to decomposition [44–46]. Silicene behaves similarly [47]. On the contrary, we find even for fully lithiated Li₆Sc₂N that the Sc—N bond length remains unaltered, which guarantees high reversibility and a long cycle life. Electron localization functions calculated for the Sc₂N and Y₂N monolayers with one adsorbed Li layer are shown in Fig. 5. We observe metallic bonding between the Li atoms, which boosts the Li diffusion at high coverage.

The charge or discharge time and power rate of a LIB depend on the electron transport and Li diffusion properties of the anode material. The reversible electrochemical reaction taking place during charge or discharge can be written as $M_2X + xLi^+ + e^- \rightleftharpoons Li_xM_2X$, with the Li⁺ ions moving through the high-band-gap liquid electrolyte between the electrodes and the electrons moving through the outer

FIG. 6. The total densities of states ($3 \times 3 \times 1$ supercell, spin-orbit coupling included) of the (a) Sc₂N and Li₂Sc₂N and (b) Y₂N and Li₂Y₂N monolayers. The Fermi energy is set to 0 eV.

circuit of the battery. Thus, excellent metallicity and fast Li diffusion are key features of an efficient electrode material. Figure 6 shows the total densities of states obtained for the Sc₂N, Li₂Sc₂N, Y₂N, and Li₂Y₂N monolayers, demonstrating metallicity with and without adsorbed Li atoms. Therefore, no metallic additives are required, which is an important advantage, as such additives lower the Li diffusion and result in experimental complications that increase the price of the LIB [13,15,46,48–50].

Figures 7(a) and 7(b) show the relative energy along two Li diffusion pathways between T sites. The trajectories of the pathways are depicted as insets. While the direct pathway (T-T) results in Li diffusion barriers of 76 and 103 meV for the Sc₂N and Y₂N monolayers, respectively, we obtain values of only 19 and 27 meV for the minimum energy pathway (T-H-T). These values are one order of magnitude lower than reported for graphene and its derivatives [9–11], silicene [14], transition-metal monochalcogenides [51], and transition-metal dichalcogenides [15,52]. They are even lower than reported for most MXenes [25,27,53]. We observe no significant variation in the monolayer thicknesses and *M*—*X* bond lengths during Li diffusion, which contributes to the low Li diffusion barriers. The smooth potential surface due to avoided

formation of ionic *M*—Li bonds (Fig. 5) promotes the Li diffusion and gives rise to superior anode materials for LIBs.

IV. CONCLUSIONS

Sc₂B, Sc₂N, Y₂B, Y₂N, and Y₂P monolayers are predicted to be stable two-dimensional materials with hexagonal symmetry. All turn out to combine high stiffness with excellent metallicity. The Sc₂N and Y₂N monolayers provide Li binding energies suitable for application in LIBs. In particular, the high Li storage capacity (1547 mAh/g) and low Li diffusion barrier (19 meV) of the Sc₂N monolayer outperform competing anode materials including graphite (372 mAh/g [54], 0.48 eV [55]). Low Li diffusion barriers enable fast charge or discharge and high power rate, while negligible structural changes during lithiation and delithiation enable high reversibility and a long cycle life. In addition, low average open-circuit voltages pave the way for high-voltage LIBs. Overall, the remarkable electrochemical properties of the Sc₂N and Y₂N monolayers point to outstanding potential as anode materials in future LIB technology.

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FIG. 7. The relative energies for Li diffusion along the pathways (a) T-H-T and (b) T-T on the Sc₂N and Y₂N monolayers. The insets show the trajectories of the pathways in top and side views.

- [1] G. Zubi, R. Dufo-López, M. Carvalho, and G. Pasaoglu, The lithium-ion battery: State of the art and future perspectives, *Renewable Sustainable Energy Rev.* **89**, 292 (2018).
- [2] T. Kim, W. Song, D.-Y. Son, L. K. Ono, and Y. Qi, Lithium-ion batteries: Outlook on present, future, and hybridized technologies, *J. Mater. Chem. A* **7**, 2942 (2019).
- [3] F. Wu, J. Maier, and Y. Yu, Guidelines and trends for next-generation rechargeable lithium and lithium-ion batteries, *Chem. Soc. Rev.* **49**, 1569 (2020).
- [4] D. Larcher, S. Beattie, M. Morcrette, K. Edstroem, J.-C. Jumas, and J.-M. Tarascon, Recent findings and prospects in the field of pure metals as negative electrodes for Li-ion batteries, *J. Mater. Chem.* **17**, 3759 (2007).
- [5] S. Goriparti, E. Miele, F. De Angelis, E. Di Fabrizio, R. P. Zaccaria, and C. Capiglia, Review on recent progress of nanostructured anode materials for Li-ion batteries, *J. Power Sources* **257**, 421 (2014).
- [6] D. Deng, Li-ion batteries: Basics, progress, and challenges, *Energy Sci. Eng.* **3**, 385 (2015).
- [7] Y. Lu, L. Yu, and X. W. D. Lou, Nanostructured conversion-type anode materials for advanced lithium-ion batteries, *Chem* **4**, 972 (2018).
- [8] H. Yildirim, A. Kinaci, Z.-J. Zhao, M. K. Y. Chan, and J. P. Greeley, First-principles analysis of defect-mediated

- Li adsorption on graphene, *ACS Appl. Mater. Interfaces* **6**, 21141 (2014).
- [9] S. Huang, Y. Li, Y. Feng, H. An, P. Long, C. Qin, and W. Feng, Nitrogen and fluorine co-doped graphene as a high-performance anode material for lithium-ion batteries, *J. Mater. Chem. A* **3**, 23095 (2015).
- [10] L. Zhou, Z. F. Hou, B. Gao, and T. Frauenheim, Doped graphenes as anodes with large capacity for lithium-ion batteries, *J. Mater. Chem. A* **4**, 13407 (2016).
- [11] A. Samad, H. J. Kim, and Y.-H. Shin, Structure stability and high Li storage capacity of the unzipped graphene oxide monolayer, *Appl. Surf. Sci.* **475**, 151 (2019).
- [12] J. Sun, H.-W. Lee, M. Pasta, H. Yuan, G. Zheng, Y. Sun, Y. Li, and Y. Cui, A phosphorene-graphene hybrid material as a high-capacity anode for sodium-ion batteries, *Nat. Nanotechnol.* **10**, 980 (2015).
- [13] G.-C. Guo, D. Wang, X.-L. Wei, Q. Zhang, H. Liu, W.-M. Lau, and L.-M. Liu, First-principles study of phosphorene and graphene heterostructure as anode materials for rechargeable Li batteries, *J. Phys. Chem. Lett.* **6**, 5002 (2015).
- [14] L. Shi, T. S. Zhao, A. Xu, and J. B. Xu, *Ab initio* prediction of a silicene and graphene heterostructure as an anode material for Li- and Na-ion batteries, *J. Mater. Chem. A* **4**, 16377 (2016).
- [15] A. Samad and Y.-H. Shin, MoS_2/VS_2 nanocomposite as a superior hybrid anode material, *ACS Appl. Mater. Interfaces* **9**, 29942 (2017).
- [16] N. Khossossi, V. Shukla, Y. Benhouria, I. Essaoudi, A. Ainane, R. Ahuja, G. Babu, and P. M. Ajayan, Exploring the possibility of β -phase arsenic-phosphorus polymorph monolayer as anode materials for sodium-ion batteries, *Adv. Theory Simul.* **3**, 2000023 (2020).
- [17] M. Sun and U. Schwingenschlögl, B_2P_6 : A two-dimensional anisotropic Janus material with potential in photocatalytic water splitting and metal-ion batteries, *Chem. Mater.* **32**, 4795 (2020).
- [18] B. Akgenc, Two-dimensional black arsenic for Li-ion battery applications: A DFT study, *J. Mater. Sci.* **54**, 9543 (2019).
- [19] F. Ersan, G. Gökoğlu, and E. Aktürk, Adsorption and diffusion of lithium on monolayer transition metal dichalcogenides ($\text{MoS}_2(1-x)\text{Se}_2x$) alloys, *J. Phys. Chem. C* **119**, 28648 (2015).
- [20] F. Ersan, H. D. Ozaydin, G. Gökoğlu, and E. Aktürk, Theoretical investigation of lithium adsorption, diffusion and coverage on MX_2 ($M = \text{Mo}, \text{W}$; $X = \text{O}, \text{S}, \text{Se}, \text{Te}$) monolayers, *Appl. Surf. Sci.* **425**, 301 (2017).
- [21] Y. Du, X. Zhu, L. Si, Y. Li, X. Zhou, and J. Bao, Improving the anode performance of WS_2 through a self-assembled double carbon coating, *J. Phys. Chem. C* **119**, 15874 (2015).
- [22] H. Shu, F. Li, C. Hu, P. Liang, D. Cao, and X. Chen, The capacity fading mechanism and improvement of cycling stability in MoS_2 -based anode materials for lithium-ion batteries, *Nanoscale* **8**, 2918 (2016).
- [23] R. Meshkian, L.-Å. Näslund, J. Halim, J. Lu, M. W. Barsoum, and J. Rosen, Synthesis of two-dimensional molybdenum carbide, Mo_2C , from the gallium based atomic laminate $\text{Mo}_2\text{Ga}_2\text{C}$, *Scripta Mater.* **108**, 147 (2015).
- [24] D. Çakır, C. Sevik, O. Gülseren, and F. M. Peeters, Mo_2C as a high capacity anode material: A first-principles study, *J. Mater. Chem. A* **4**, 6029 (2016).
- [25] Q. Sun, Y. Dai, Y. Ma, T. Jing, W. Wei, and B. Huang, *Ab initio* prediction and characterization of Mo_2C monolayer as anodes for lithium-ion and sodium-ion batteries, *J. Phys. Chem. Lett.* **7**, 937 (2016).
- [26] G. Gao, G. Ding, J. Li, K. Yao, M. Wu, and M. Qian, Monolayer MXenes: Promising half-metals and spin gapless semiconductors, *Nanoscale* **8**, 8986 (2016).
- [27] A. Samad, A. Shafique, H. J. Kim, and Y.-H. Shin, Superionic and electronic conductivity in monolayer W_2C : *Ab initio* predictions, *J. Mater. Chem. A* **5**, 11094 (2017).
- [28] B.-M. Jun, S. Kim, J. Heo, C. M. Park, N. Her, M. Jang, Y. Huang, J. Han, and Y. Yoon, Review of MXenes as new nanomaterials for energy storage/delivery and selected environmental applications, *Nano Res.* **12**, 471 (2019).
- [29] Y. Yu, J. Zhou, and Z. Sun, Modulation engineering of 2D MXene-based compounds for metal-ion batteries, *Nanoscale* **11**, 23092 (2019).
- [30] T. Zhang, D. Li, Z. Tao, and J. Chen, Understanding electrode materials of rechargeable lithium batteries via DFT calculations, *Prog. Nat. Sci.* **23**, 256 (2013).
- [31] A. Urban, D.-H. Seo, and G. Ceder, Computational understanding of Li-ion batteries, *npj Comput. Mater.* **2**, 16002 (2016).
- [32] Q. He, B. Yu, Z. Li, and Y. Zhao, Density functional theory for battery materials, *Energy Environ. Mater.* **2**, 264 (2019).
- [33] G. Kresse and D. Joubert, From ultrasoft pseudopotentials to the projector augmented-wave method, *Phys. Rev. B* **59**, 1758 (1999).
- [34] S. Grimme, J. Antony, S. Ehrlich, and H. Krieg, A consistent and accurate *ab initio* parametrization of density functional dispersion correction (DFT-D) for the 94 elements H-Pu, *J. Chem. Phys.* **132**, 154104 (2010).
- [35] H. J. Monkhorst and J. D. Pack, Special points for Brillouin-zone integrations, *Phys. Rev. B* **13**, 5188 (1976).
- [36] A. Togo and I. Tanaka, First principles phonon calculations in materials science, *Scr. Mater.* **108**, 1 (2015).
- [37] Y. Le Page and P. Saxe, Symmetry-general least-squares extraction of elastic data for strained materials from *ab initio* calculations of stress, *Phys. Rev. B* **65**, 104104 (2002).
- [38] G. Henkelman, B. P. Uberuaga, and H. Jónsson, A climbing image nudged elastic band method for finding saddle points and minimum energy paths, *J. Chem. Phys.* **113**, 9901 (2000).
- [39] X. Lv, W. Wei, Q. Sun, L. Yu, B. Huang, and Y. Dai, Sc_2C as a promising anode material with high mobility and capacity: A first-principles study, *ChemPhysChem* **18**, 1627 (2017).
- [40] M. Born and K. Huang, *Dynamical Theory of Crystal Lattices* (Clarendon Press, Oxford, 1954).
- [41] See the Supplemental Material at <http://link.aps.org/supplemental/10.1103/PhysRevApplied.15.034025> for the structures of bulk Y_2N and Sc_2N with adsorbed Li layers.
- [42] H. Wang, E. Liu, Y. Wang, B. Wan, C.-H. Ho, F. Miao, and X. Wan, Cleavage tendency of anisotropic two-dimensional

- materials: Re X₂ (X = S, Se) and WTe₂, *Phys. Rev. B* **96**, 165418 (2017).
- [43] Q. Tang, Z. Zhou, and P. Shen, Are MXenes promising anode materials for Li ion batteries? Computational studies on electronic properties and Li storage capability of Ti₃C₂ and Ti₃C₂X₂ (X = F, OH) monolayer, *J. Am. Chem. Soc.* **134**, 16909 (2012).
- [44] A. Samad, A. Shafique, and Y.-H. Shin, Adsorption and diffusion of mono, di, and trivalent ions on two-dimensional TiS₂, *Nanotechnology* **28**, 175401 (2017).
- [45] L. David, R. Bhandavat, and G. Singh, MoS₂/graphene composite paper for sodium-ion battery electrodes, *ACS Nano* **8**, 1759 (2014).
- [46] A. Samad, M. Noor-A-Alam, and Y.-H. Shin, First principles study of a SnS₂/graphene heterostructure: A promising anode material for rechargeable Na ion batteries, *J. Mater. Chem. A* **4**, 14316 (2016).
- [47] G. A. Tritsarlis, E. Kaxiras, S. Meng, and E. Wang, Adsorption and diffusion of lithium on layered silicon for Li-ion storage, *Nano Lett.* **13**, 2258 (2013).
- [48] J. Lin, Z. Peng, C. Xiang, G. Ruan, Z. Yan, D. Natelson, and J. M. Tour, Graphene nanoribbon and nanostructured SnO₂ composite anodes for lithium ion batteries, *ACS Nano* **7**, 6001 (2013).
- [49] Y. Fang, Y. Lv, F. Gong, A. A. Elzatahry, G. Zheng, and D. Zhao, Synthesis of 2D-mesoporous-carbon/MoS₂ heterostructures with well-defined interfaces for high-performance lithium-ion batteries, *Adv. Mater.* **28**, 9385 (2016).
- [50] Y.-T. Du, X. Kan, F. Yang, L.-Y. Gan, and U. Schwingenschlögl, MXene/graphene heterostructures as high-performance electrodes for Li-ion batteries, *ACS Appl. Mater. Interfaces* **10**, 32867 (2018).
- [51] S. Karmakar, C. Chowdhury, and A. Datta, Two-dimensional group IV monochalcogenides: Anode materials for Li-ion batteries, *J. Phys. Chem. C* **120**, 14522 (2016).
- [52] D. Wang, Y. Liu, X. Meng, Y. Wei, Y. Zhao, Q. Pang, and G. Chen, Two-dimensional VS₂ monolayers as potential anode materials for lithium-ion batteries and beyond: First-principles calculations, *J. Mater. Chem. A* **5**, 21370 (2017).
- [53] D. Er, J. Li, M. Naguib, Y. Gogotsi, and V. B. Shenoy, Ti₃C₂ MXene as a high capacity electrode material for metal (Li, Na, K, Ca) ion batteries, *ACS Appl. Mater. Interfaces* **6**, 11173 (2014).
- [54] K. Persson, Y. Hinuma, Y. S. Meng, A. Van der Ven, and G. Ceder, Thermodynamic and kinetic properties of the Li-graphite system from first-principles calculations, *Phys. Rev. B* **82**, 125416 (2010).
- [55] K. Toyoura, Y. Koyama, A. Kuwabara, F. Oba, and I. Tanaka, First-principles approach to chemical diffusion of lithium atoms in a graphite intercalation compound, *Phys. Rev. B* **78**, 214303 (2008).