The Comparison of Intercalation of Li Atoms in SnS_2 Anode of Battery and $TaSe_2$ Anode of Battery: ab initio Calculation

Prikhozha Yu.O., Balabai R.M.

Kryvyi Rih State Pedagogical University, 54 Gagarina Ave., Kryvyi Rih-50086, Ukraine

E-mail: prihozhaya.yuliya93@gmail.com, balabai@i.ua

Introduction and Statement of the Problem

Motivated by the broad need to develop battery-based energy storage technologies, ion intercalation into materials suitable for these applications has been pursued intensively in recent years. Layered transition metal dichalcogenides featuring a Van der Waals interlayer coupling are capable of hosting ions in its various interstitial sites, making transition metal dichalcogenides an important class of electronic materials for ion intercalation studies [1]. In all these studies, however, the process of Li ion intercalation and its effect on the electrical transport in the thin crystal were not studied in detail [2-3]. In order to minimise the disadvantages in the manufacture for anode materials, it is necessary to use in advance a computational experiment, which is based on quantum calculation from the ab initio.

Methods and Models of Calculations

The calculated results were obtained using the author`s program code [4]. The basic states of the electron-nucleus systems were detected by means of the self-consistent solution of the Kohn-Sham equations. To reproduce the material of the battery anode for the accumulation of Li atoms in the form of two endless monolayers SnS_2 or $TaSe_2$ the orthorhombic type artificial superlattice was created ($a \neq b \neq c, \alpha = \beta = \gamma = 90°$). The research object determined parameters of the superlattice and the atomic base. For simulation of the film structure, the lattice parameter cell in the direction of the ***a***, ***b***, crystallographic axis were modeled the infinite SnS_2 film or $TaSe_2$ film, in a plane perpendicular to the surface the ***c*** crystallographic axis was chosen so that the translationally located films would not influence one another. Li atoms were placed according their location in solid state at low temperatures. Algorithm for calculating the energy barriers of migration of Li atoms was such that for each atomic configuration that corresponded to the elementary step of spatial migration, the total energy was calculated be the formula (1) thus generating the energy relief along the migration trajectory of Li atoms. The reference level of energy on the energy relief graphs was taken to be equal to the value of the total energy of the system at zero filling of the layer SnS_2 or $TaSe_2$ (fig. 1).

$$\frac{E_{total}}{\Omega} = \sum_{\vec{k},\vec{G},i} \left|b_i(\vec{k}+\vec{G})\right|^2 \frac{\hbar^2}{2m}(\vec{k}+\vec{G})^2 + \frac{1}{2}4\pi e^2 \sum_{\vec{G}} \frac{`\left|\rho(\vec{G})\right|^2}{\vec{G}^2} + \sum_{G} \varepsilon_{xc}(\vec{G})\,\rho^*(\vec{G}) + \sum_{\vec{k},\vec{G},\vec{G}`,i,l,s} S_s(\vec{G}-\vec{G}`)\Delta V_{l,s}^{NL}(\vec{k}+\vec{G},\vec{k}+\vec{G}`)b_i\left(\vec{k}+\overrightarrow{G})b_i^*(\vec{k}+\vec{G}`) + \sum_{\vec{G},s} `S_s(\vec{G}\right)V_s^L(\vec{G})\rho^*(\vec{G}) + \left\{\sum_s \alpha_s\right\}\left[\Omega^{-1}\sum_s Z_s\right] + \Omega^{-1}\gamma_{Ewald}$$

Where $\vec{k}$ is of the first Brillouin zone, $\vec{G}$ is the vector of the reciprocal lattice, $\Psi_i(\vec{k}+\vec{G})$ is the coefficient of the expansion of the wave function, i denotes occupied states for a certain $\vec{k}$, $\rho(\vec{G})$ is the coefficient of decay of the density of valence electrons, s number atoms in an elementary cell, $S_s(\vec{G})$ is a structural factor, V_S^L is a local (l-independent) spherically symmetric pseudopotential, l denotes the quantum orbital number, $\Delta V_{l,\vec{\tau}}^{NL}$ is a nonlocal (l-dependent) additive to V_S^L, Z_S is an ion charge, γ_{Ewald} is the Madelung energy of points ions.

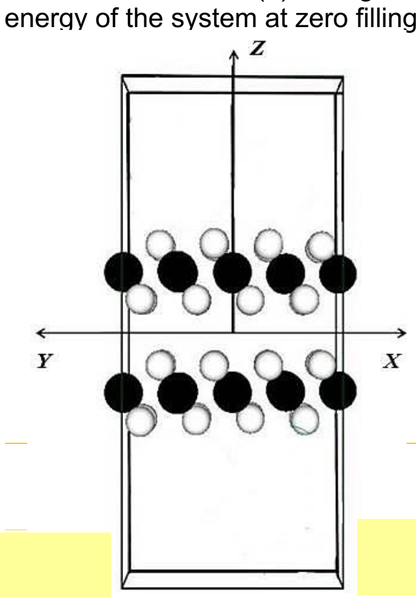

Fig.1. The direction of migration of Li atoms from each other into the interlayer of SnS_2 or TaSe2 layer within the cell, view of a primitive cell with an atomic basis

Results and Discussion

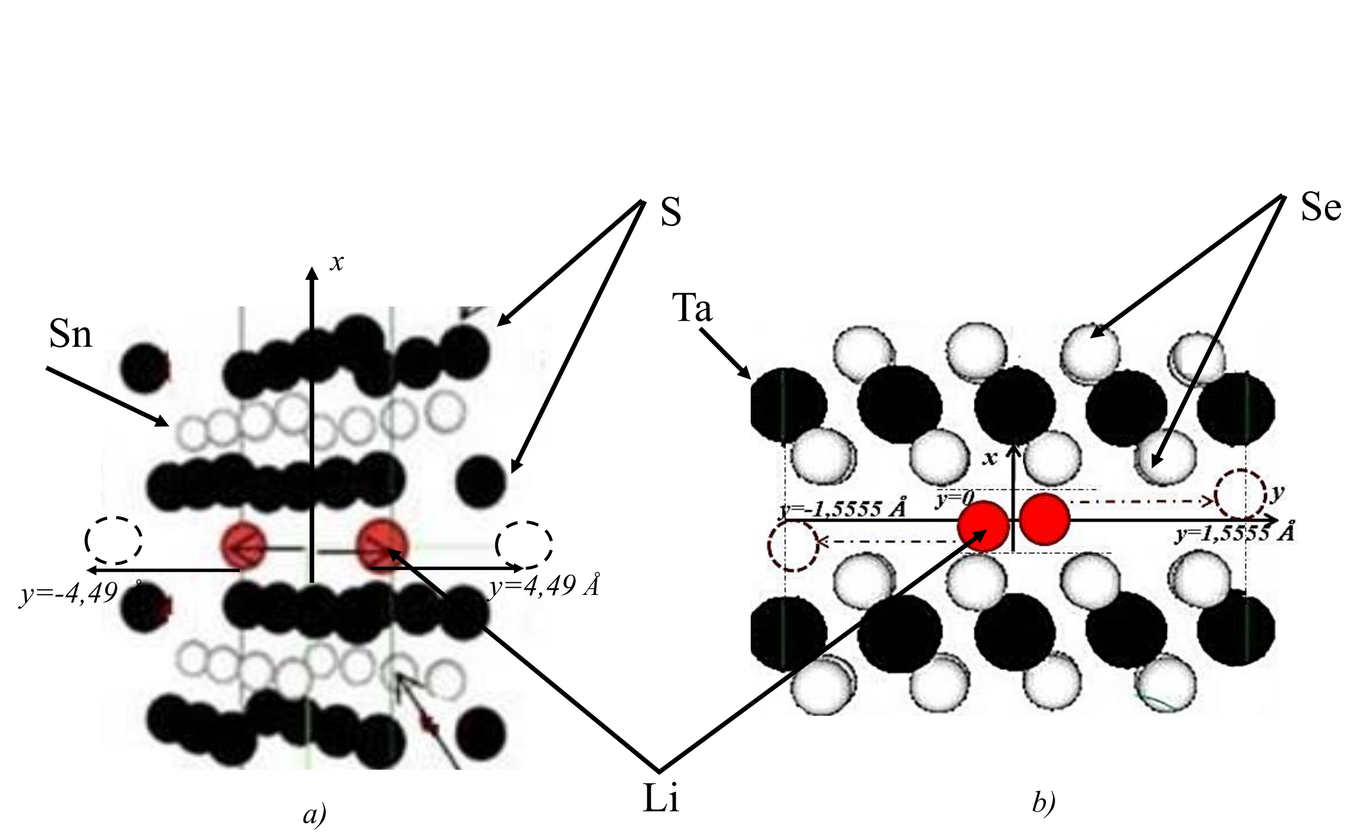

Fig. 2. The trajectory and direction of movement of intercalated Li atoms in the interlayer layers of anodes made of films SnS_2 (a); the trajectory and direction of movement of intercalated Li atoms in the interlayer layers of anodes made of films $TaSe_2$ (b)

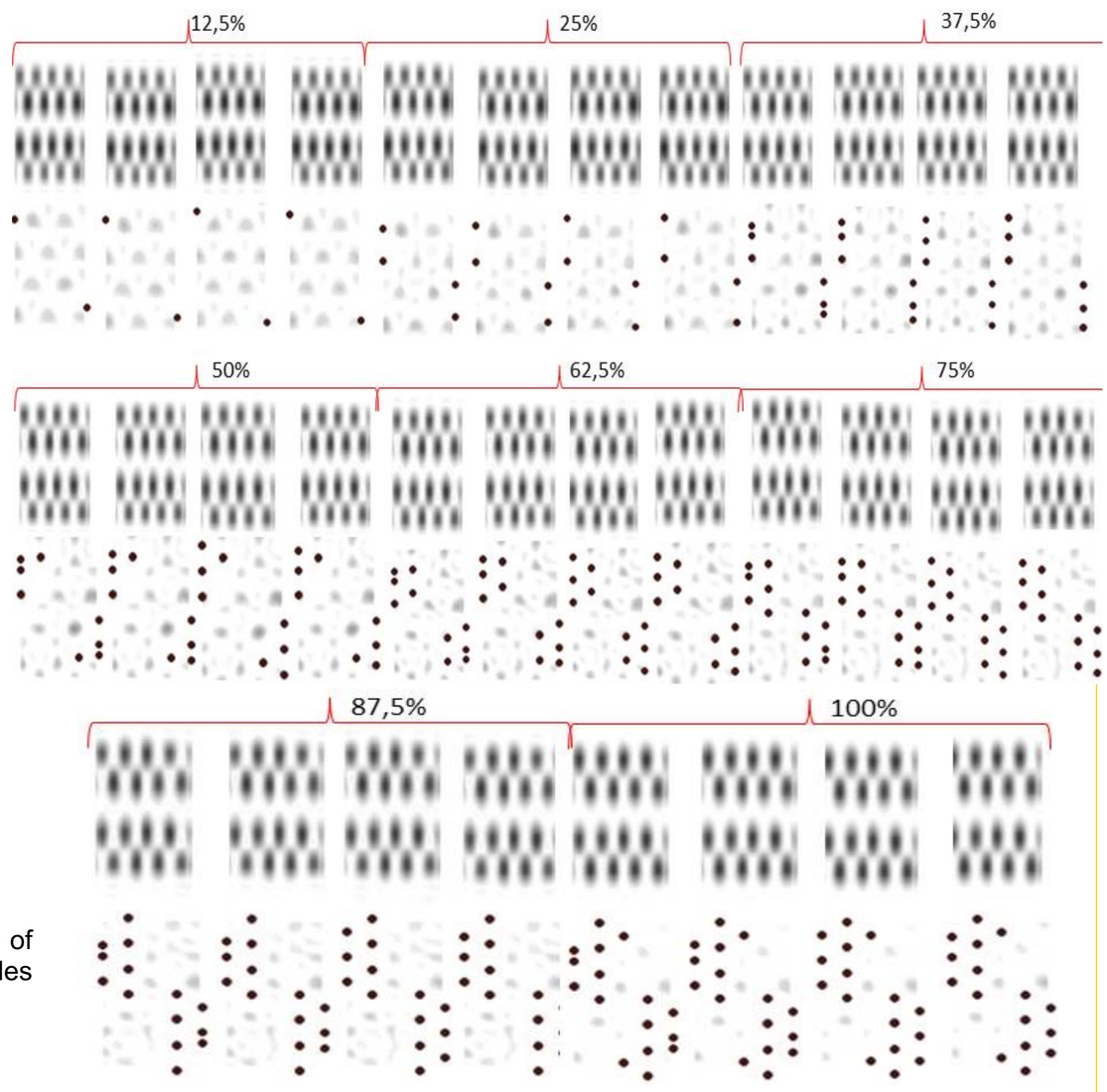

Fig. 3. Sections of the [110] and [100] spatial distributions of valence electron density within the primitive cell for the filling of the SnS2 interlayer with Li atoms at filling levels from 12.5% to 100% at different positions of Li atoms from y=3.05342 Å to y=4.13108 Å from left to right

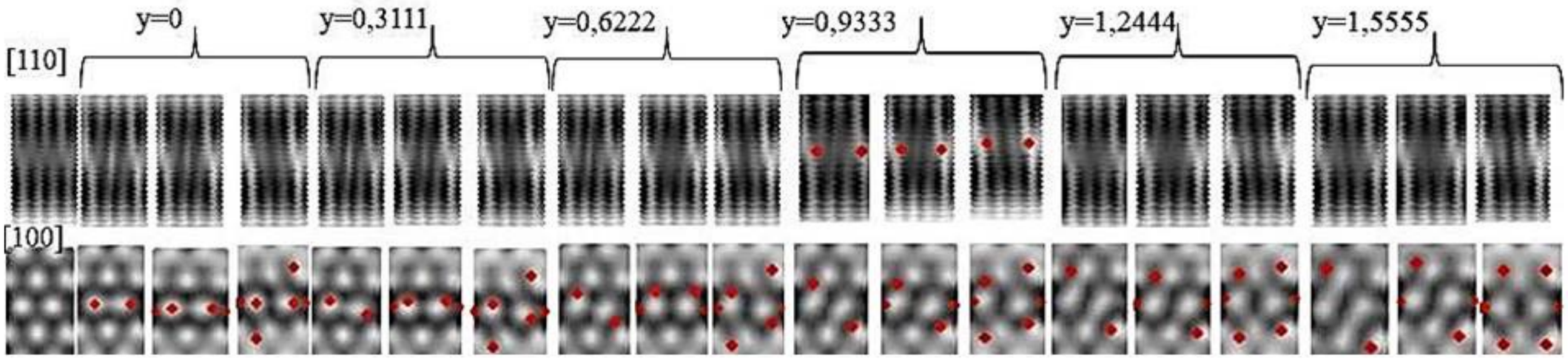

Fig. 4. Fig.6. The cross sections [110] and [100] of the spatial distributions of the valence electron density within a cell whose atomic basis simulates two $TaSe_2$ monolayers, with Li atoms intercalated into the interlayer space, at 33,3%, 66,7%, 100% atoms for y=0 – 1.5555 Å

Conclusions

An increase in the value of the energy barrier was recorded with an increase in the number of intercalated Li atoms filling the interlayer space of anode materials SnS_2 and $TaSe_2$.

References

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