Effects of Continuous Chalcogen Substitution on Energetics and Electronic Properties of WSSe

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Transition metal dichalcogenides (TMDs) can form in-plane and out-of-plane heterostructures whose physical properties are sensitive to their morphologies [1]. Among such heterostructures, Janus TMDs are now keeping a premier position in the field of atomic layer materials because they have unique heterostructure across their atomic layers: A transition metal layer is sandwiched by different chalcogen atomic layers leading to peculiar electronic structure owing to the internal electric field [2]. Experiments reported that Janus TMDs can be obtained by sulfurization or selenization on MSe_2 or MS_2, where M is a transition metal, respectively [3]. However, the detailed mechanism of Janus TMD formation is still unclear. Thus, this work aims to clarify the formation mechanism of Janus WSSe by conducting the first-principles total energy calculations based on the density functional theory with the effective screening medium method.
Our calculations on $WS_xSe_{(1-x)}$ demonstrated that WS_2 is the most stable structure. In contrast, Janus WSSe is the least stable structure. In addition, during the sulfurization from WSe_2 to WSSe, $WS_{0.555}Se_{1.444}$ is the metastable structure possessing the lowest total energy because of the competition between the energy cost of dipole energy and the energy gain of S-W bond formation. The electronic structure of $WS_xSe_{(1-x)}$ shows contentious change concerning sulfur concentration, inconsistent with an in-situ observation of Janus formation. Therefore, our calculations imply that contentious sulfurization is not a possible synthesis process for the Janus formation [3].

References

[1] M. Maruyama et al., ACS Appl. Nano Mater. 6 (2023) 5434.
[2] Y. Gao, S. Okada, Appl. Phys. Express 16 (2003) 075004.
[3] Y. Gao, M. Maruyama, S. Okada, Jpn. J. Appl. Phys. 63 (2024) 065001.

Figure

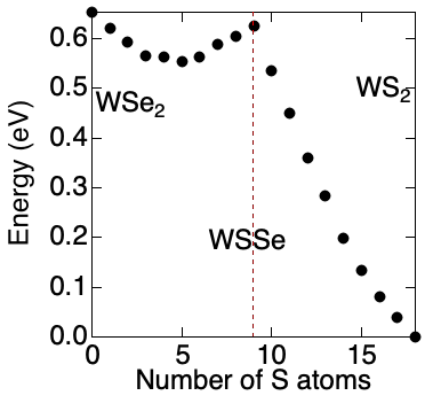

Figure 1: Relative total energy per W atom of $WS_{(2-x)}Se_x$ as a function of the number of S atoms n. WSe_2, WS_2, and Janus WSSe correspond to n = 0, 18, and 9, respectively. The energies are measured based on that of WS_2.