Development of an approach to the construction of phase diagrams of materials with metallic and covalent bond types within the CALPHAD on the basis of experimental data and results of atomistic modeling using the example of the Ti-Si-H system.

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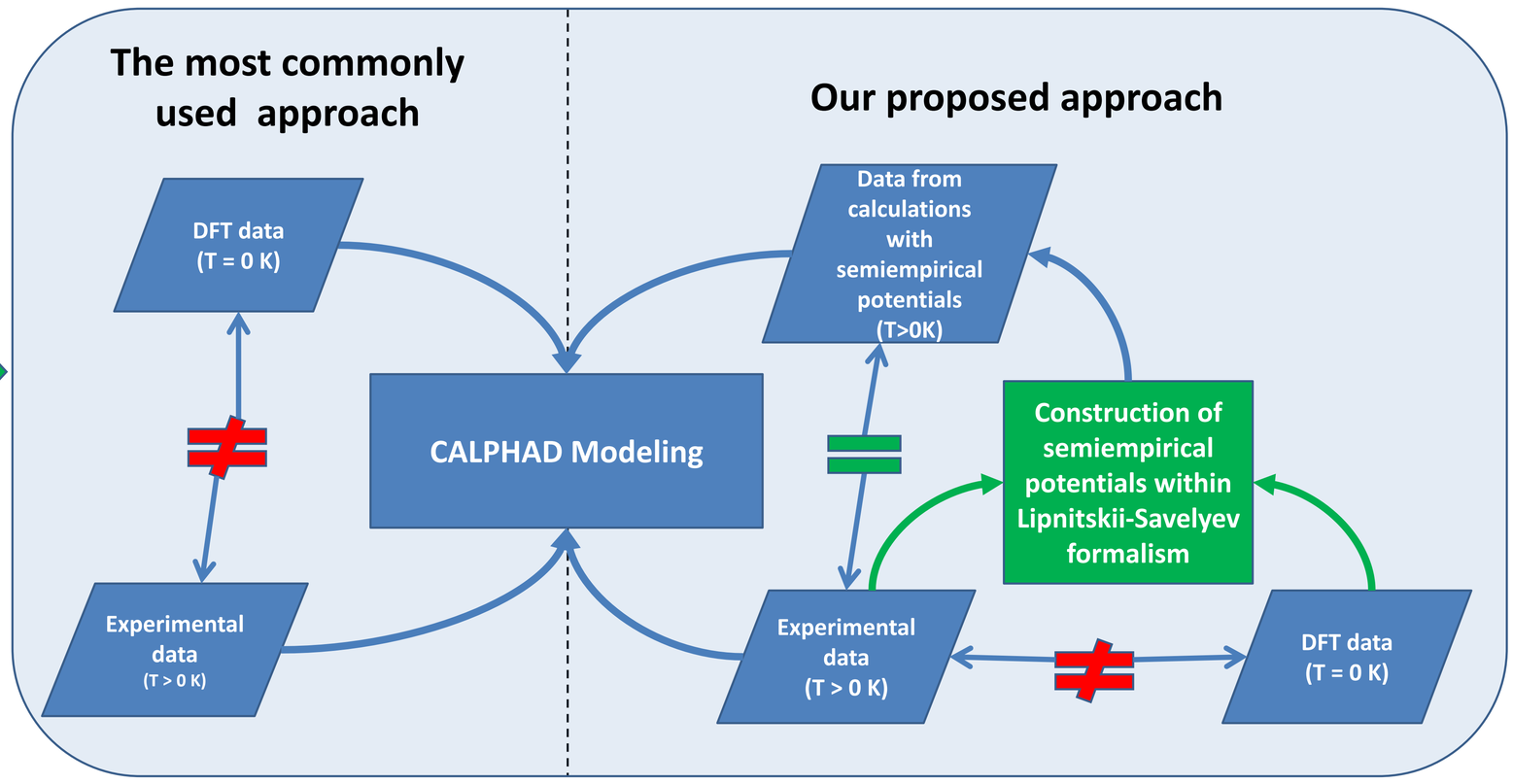
Formulation of the problem

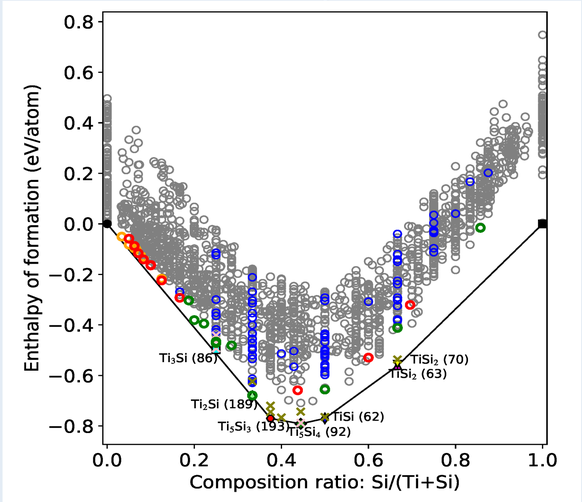
Construction and prediction of phase diagrams of substances in the framework of the CALPHAD (CALculation of PHAse Diagrams) approach [1] is one of the key components of the optimization of the development of new materials. However, CALPHAD does not allow to predict the existence of phases with structures not included in databases. This predictive task is successfully resolved by a combination of the high-throughput DFT (Density Functional Theory) calculations with data mining techniques and genetic algorithms [2]. **The problem is in the connection** between the DFT based predictions and CALPHAD models based on the experimental data.

The reasons for this problem include the limitations of DFT calculations at high temperatures and large atomic scales, and model representations of the exchange-correlation energy functional, the most common of which are GGA and LDA approximations, which not rarely leads to inconsistency of DFT results with experimental data.



Obtained results

Results of the evolutionary search for new compounds in the Ti–Si system



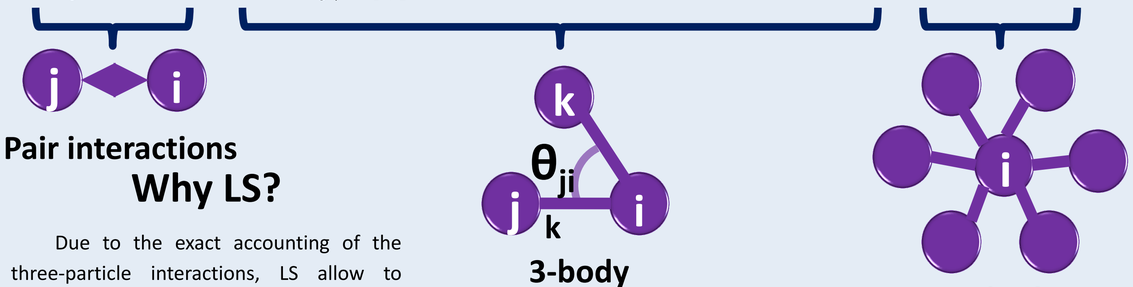
Why Ti-Si-H?

Ti-Si-H is a fairly representative system containing a metal, a semiconductor and a light element. In this system, the compounds with metallic and covalent chemical bonds are formed. The combination of these three types of elements is found in many materials, including alloys, materials for electronics, energy storage and composite materials. The methods developed on the example of this system can be used to build interatomic potentials, calculate thermodynamic characteristics, and simulate processes in a wide range of practically important materials

The Lipnitskii-Savelyev (LS) formalism to the construction of semiempirical interatomic potentials [3]:

$$E_{tot} = \sum_{i<j}^{N} \Phi(R_{ji}) + \sum_{i}^{N} \sum_{k<j\neq i}^{N} \sum_{p,q}^{n_3} g^{pq}(cos(\theta_{jik})) \times f^{p}(R_{ji}) f^{q}(R_{ki}) + \sum_{i}^{N} F(\bar{\rho}_i)$$

Basis functions



Pair interactions

3-body interactions

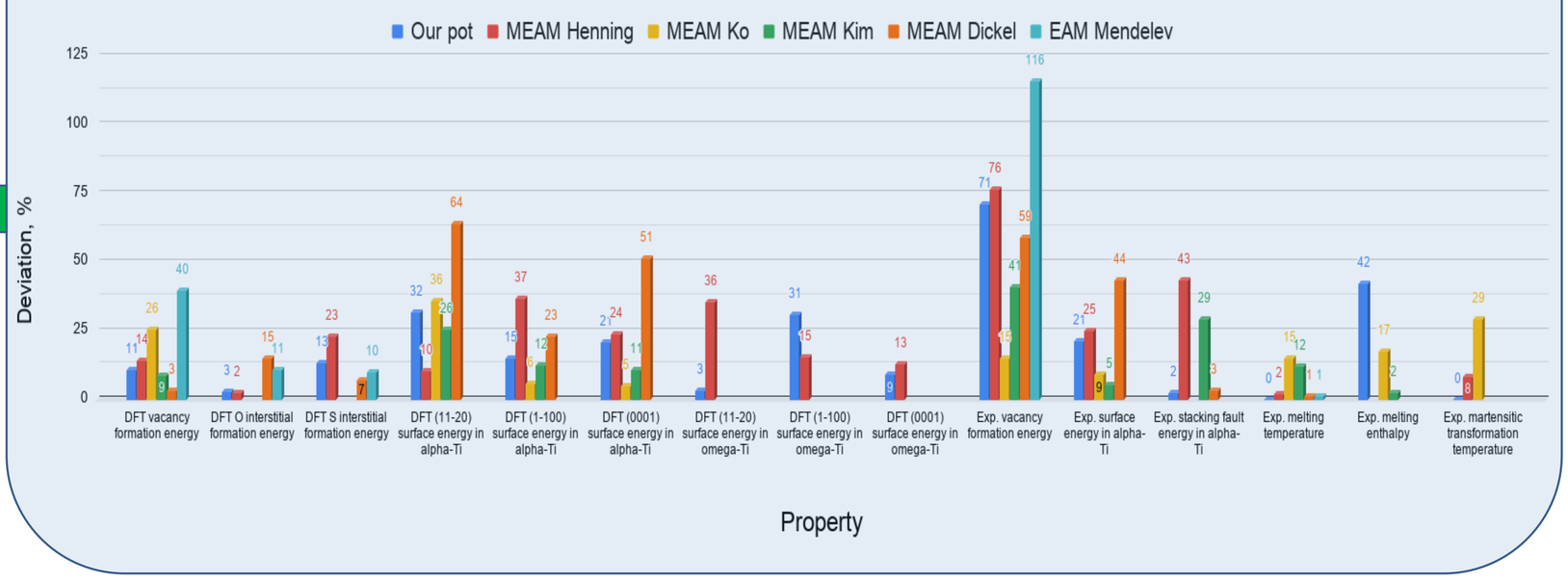
n-body interactions

Why LS?

Due to the exact accounting of the three-particle interactions, LS allow to obtain a high-precision prediction of the properties of materials with mixed types of bonding.

Thermodynamic description of the Ti-Si-H system with ab initio predicted phases

Constructed potentials for the Ti-Si-H system within the LS formalism [4]



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[4] Kartamyshev, A. I., Lipnitskii, A. G., Saveliev, V. N., Maksimenko, V. N., Nelasov, I. V., Poletaev, D. O. (2019). // Comp. Mat. Sci., 160, 30–41.