Second Principles Density Functional Theory models: a procedure to obtain their tight-binding parameters automatically.

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First-principles calculations allow to compute the energy and properties of a compound from essential information about its structure and composition. However, such simulations at operating conditions (finite temperatures or electric fields) remain almost limited by computational resources to very small length scales (a few hundreds of atoms per cell) and timescales (a few picoseconds). A practical way to overcome these limitations is to work with effective atomistic models, integrating out the electronic degrees of freedom and providing a simple parametric description of the Born-Oppenheimer energy surface in terms of structural degrees of freedom. To reintroduce explicitly the treatment of the meaningful electronic degrees of freedom, a method has been proposed in the form of a tight-binding model [1].

This tight binding approach is based on the Taylor expansion of the DFT energy around a reference electronic density. The deformation density with respect this reference, as well as the hamiltonian matrix elements, is expressed in a basis of Wannier functions. Only the relevant electrons to the problem can be retained in the description, reducing the computational cost. This gain in efficiency would come at the cost of finding the right parameters in the tight binding hamiltonian matrix elements, that would reproduce as close as possible the DFT calculations.

Here we propose a method implemented in python, the MODELMAKER code, for the automatic parametrization of such tight binding hamiltonian. The starting point is made of first-principles calculations (from the SIESTA code [2]) in training sets with a small number of atoms in the unit cell. Since there is not input coming from the experiment, our method retains full predictive power, and that is why it is coined as second-principles simulations. Both electron-lattice coupling and electron-electron interactions are included in the parametrization of the electronic model. The obtained parameters are validated comparing the band structures computed from first and second-principles in configurations which are not included in the training set.

[1] P. García-Fernández, et al; *Phys.Rev.B* **93** 195137 (2016).
[2] A. García, et al; *The Journal of Chemical Physics* **152** 204108 (2020).