# Improving crystal structure predictions using dimers of clusters cut from crystals

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## I. SAPT BASED FORCE FIELD USED IN UPACK

A manual of UPACK is available on Ref. [1]. By default UPACK accepts Lennard-Jones and Buckingham functional forms of the force fields. Whereas, SAPT based force fields developed by autoPES [2] have the following functional form:

$$V = \sum_{a\in A, b\in B} u_{ab}(r_{ab}) = V_{\rm elst} + V_{\rm exp} + V^{(2)}_{\rm asymp} = \sum_{a\in A, b\in B} \left[ u_{{\rm elst},ab}(r_{ab}) + u_{{\rm exp},ab}(r_{ab}) + u^{(2)}_{{\rm asymp},ab}(r_{ab}) \right] \tag{1}$$

where $a$ ($b$) goes over the sets of atoms in monomer A (B), respectively. The atom-atom function are of the form

$$\begin{aligned} u_{{\rm elst},ab}(r_{ab}) =& f_1(\delta^{ab}_1, r_{ab}) \frac{q_a q_b}{r_{ab}} \\ u_{{\rm exp},ab}(r_{ab}) =& \left[ 1 + \sum_{i=1}^{k} a^{ab}_i (r_{ab})^i \right] e^{\alpha^{ab} - \beta^{ab} r_{ab}} + \frac{A^{ab}_{12}}{(r_{ab})^{12}} \\ u^{(2)}_{{\rm asymp},ab}(r_{ab}) =& - \sum_{n=6,8} f_n(\delta^{ab}_n, r_{ab}) \frac{C^{ab}_n}{(r_{ab})^n}, \end{aligned} \tag{2}$$

where $f_n$ are Tang-Toennies damping functions [3]

$$f_n(\delta^{ab}_n, r_{ab}) = 1 - e^{-\delta r} \sum_{m=0}^{n} \frac{(\delta r)^m}{m!}. \tag{3}$$

The atom-atom functional form developed by autoPES is complicated compared to the default functional forms handled by UPACK. The format for reading atom-atom Lennard-Jones and Buckingham parameters is explained in detailed in Ref. [1]. To use SAPT based force field developed using autoPES in UPACK `ibuck` (see Ref. [1] for definition) should be set to 1. The format of autoPES force field used in UPACK is given as follows:

```
OA OA 4029.91 2.38 234.81 0.0 2615.00 0.037 -0.12 7.54 7.55 100
```

where,

columns 1 and 2: atoms name for which the parameters are specified.

column 3: $e^{\alpha^{ab}}$

column 4: $\beta^{ab}$

column 5: $C^{ab}_6$

column 6: $C^{ab}_8$

column 7: $A^{ab}_{12}$

column 8: $a^{ab}_1$

column 9: $a_2^{ab}$

column 10: $\delta_1^{ab}$

column 11: $\delta_6^{ab}$

column 12: $\delta_8^{ab}$

For undamped parameters, the $\delta$'s should be set to 100.

Files for performing crystal structure predictions using SAPT based force field is provided in `$HOME/upack/data/methanol`, where `$HOME` is the home directory.

[1] THE UPACK PROGRAM PACKAGE. http://www.crystal.chem.uu.nl/~vaneyck/upack.html.

[2] Metz, M. P. & Szalewicz, K. Automatic generation of flexible-monomer intermolecular potential energy surfaces. *J. Chem. Theory Comput.* **16**, 2317–2339 (2020).

[3] Tang, K. T. & Toennies, J. P. An improved simple-model for the van der Waals potential based on universal damping functions for the dispersion coefficients. *J. Chem. Phys.* **80**, 3726–3741 (1984).