

Insights on the robustness, sensitivity and expressivity of simple many-body potentials: application to α -Zr

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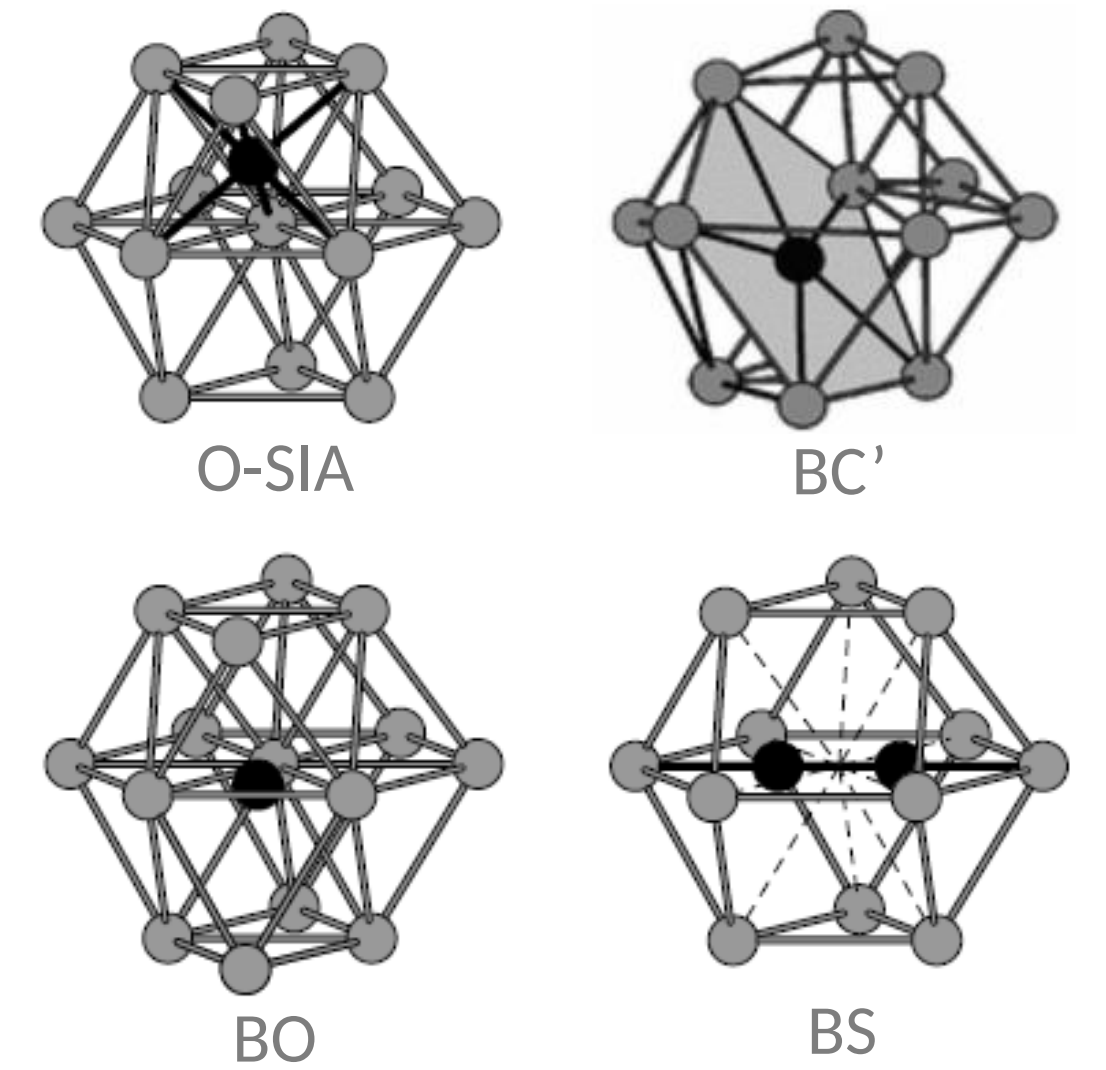
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Introduction

Interatomic potentials allow to simulate complex phenomena at length and time scales that outrun the capacity of the most accurate *ab initio* approaches. Many-body potentials, like EAM and tight binding SMA have a limited number of parameters and are based on physically-inspired functional forms, which ensures a reasonable transferability. The identification of the potential parameters for a given system is of crucial importance, but challenging.

In this work, we apply model screening and sensitivity analysis techniques to get insights on the capabilities of two SMA potentials, in the context of irradiated α -zirconium. We focus on properties that are relevant for irradiation, *i.e.* **elastic constants, point defect properties and stacking faults energies**.

The screening of the potential allows to check the robustness of the potential, *i.e.* to identify the possible artefacts related to the roughness of the potential. After correction, the most influential parameters on the computed properties are identified. In particular, one parameter affects most of the self-interstitial properties, while having a limited effect on bulk and vacancy properties. This suggests a sensitivity indices-oriented technique to refit existing potentials. Following this approach, we refine a potential for irradiation defects in α -Zr, thanks to a multi-objective optimization package.

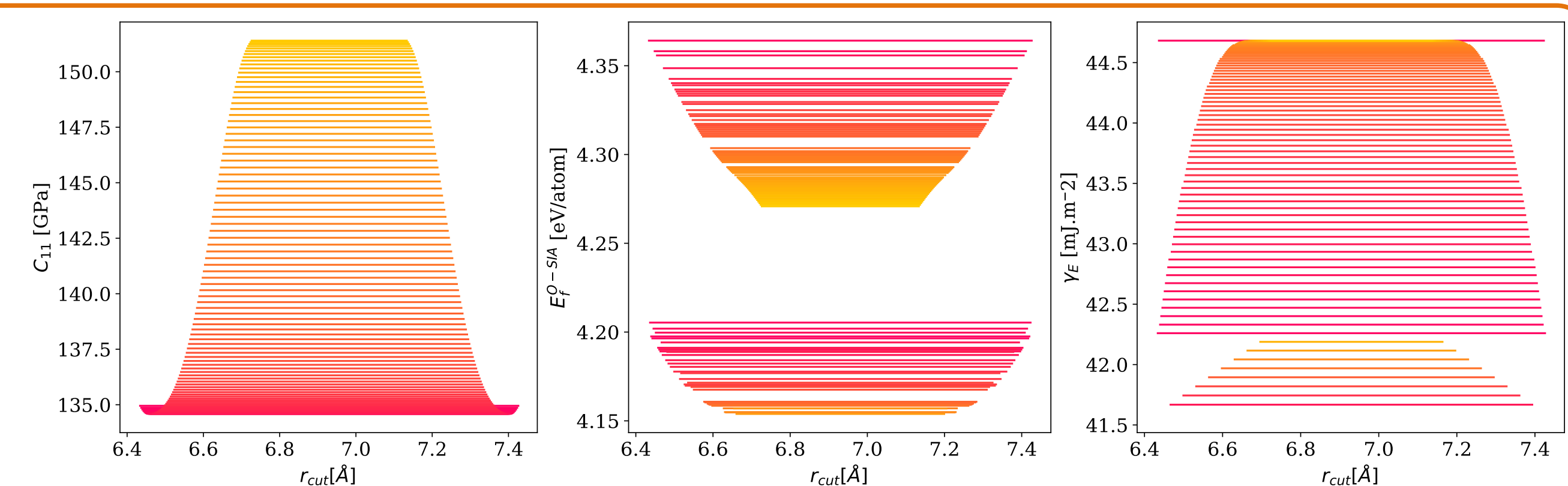
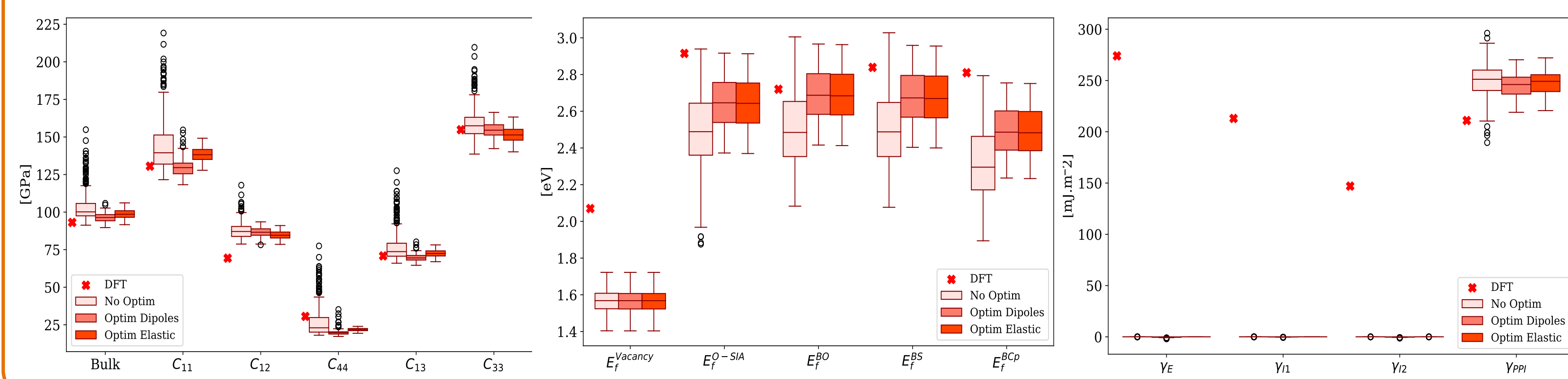


Vérité et al., Phys. Rev. B, 87 (2013)

Model screening and robustness

$$\text{TB SMA potential: } E_i^{\text{tot}} = -\xi \sum_{j \neq i}^{r_{\text{cut}}} e^{-2q \left(\frac{r_{ij}}{r_0} - 1 \right)} + A \sum_{j \neq i}^{r_{\text{cut}}} e^{-p \left(\frac{r_{ij}}{r_0} - 1 \right)}$$

- Nominal r_{cut} interval (SMA "Dufresne" potential¹), varying parameters in 2.5% range from the nominal set;
- **High spread** of observed properties, **outliers**.

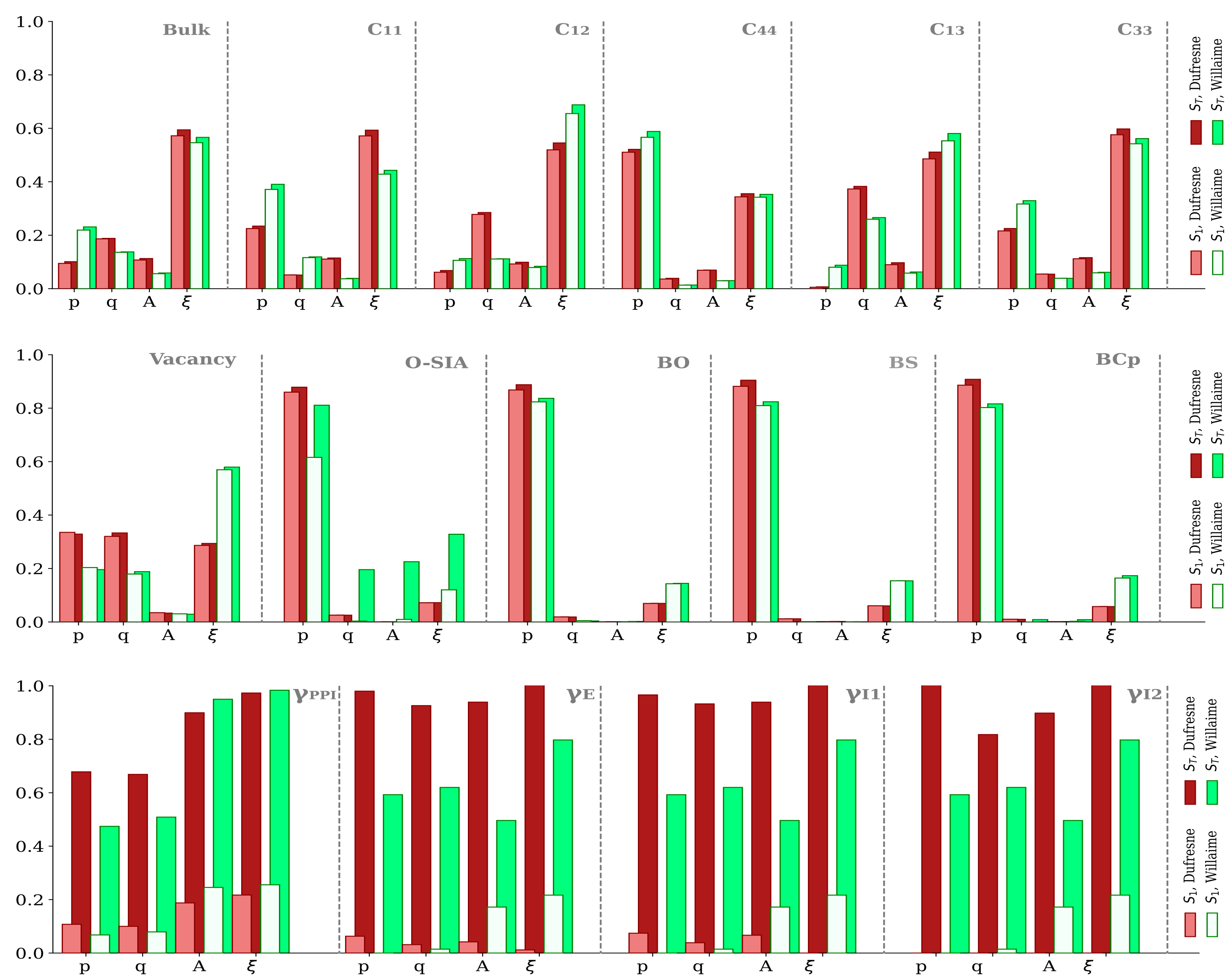


- Nominal SMA "WM1"² potential's parameters $\theta = \{A, p, \xi, q\}$;
- Cutoff radius (r_{cut}) interval varying in 30% range of the nominal case;
- Complex mapping $C_{ij} = C_{ij}(r_{\text{cut}})$, $E_f^i = E_f^i(r_{\text{cut}})$, $\gamma = \gamma(r_{\text{cut}})$;

- Optimization of the cutoff radius based on the **convergence of C_{ij} and elastic dipoles** of point defects;
- Reduction of artefacts / improved robustness: suppression of outliers, modification of median values, reduces asymmetry.

¹Dufresne et al., J. Phys. Condens. Matter, 27 (2015) ²Willaime et al., Phys. Rev. B, 43 (1991)

Identification of the most influential parameters



- Sobol'³ sensitivity indices quantify the part of the variance of the observed properties due to the variance of the parameters of the potential;
- First-order indices S_i : $S_i = V_i/V_y$;
- Higher-order indices S_{ij} : $S_{ij} = V_{ij}/V_y$
- Total indices S_{Ti} quantify the total effect of parameters on the observed property: $S_T = S_i + S_{ij} + S_{ijk} + \dots + S_{i_1 \dots i_k}$

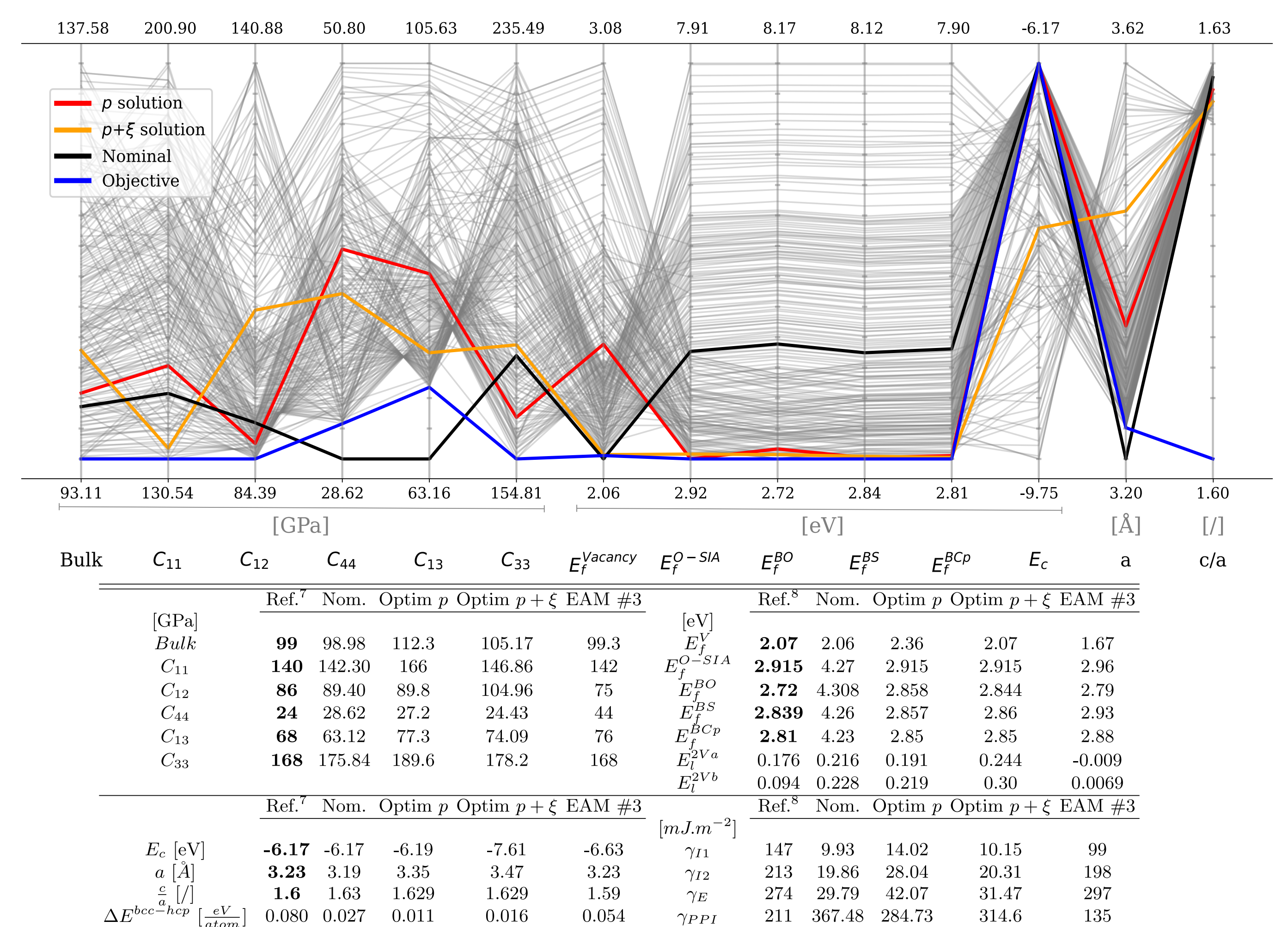
Property	S_p	S_q	S_A	S_ξ	$S_{p,q}$
E_f^{vacancy}	0.27				
$E_f^{\text{O-SIA}}$	0.34	0.38			
γ_{PI}	0.39	0.36			0.08

- p : strong effect on E_f^{SIA} , moderate effect on E_f^{vacancy} and C_{ij} (except C_{44});
- ξ : strong effect on C_{ij} , rather important on E_f^{vacancy} ;
- γ_{SF} : effects of parameters are indirect. fairly high influence of (p, ξ) interaction.

³Sobol', Math. Model. Comput. Exp., 407 (1993)

Sensitivity indices-oriented refit of the "Willaime" potential

- Refitting using only p and (p, ξ) couple, A and q are kept nominal;
- PCE⁴ surrogate model of observed properties;
- $p = [0.42, 3]$, $\xi = [7, 13.94]$, $A = 0.17936$, $q = 2.1$;
- Multi-objective optimization algorithm NSGA-II⁵ (pymoo⁶);
- 14 DFT objectives (6 C_{ij} , 5 E_f^i , 2 lattice parameters a and c/a and cohesive energy E_c);
- **Improvement** of the formation energies E_f^i of point defects and C_{44} ;
- Little **degradation** of C_{ij} , lattice parameter a and cohesive energy E_c ;
- **Stacking fault energies not well reproduced** -> link with c/a ratio and expressivity limit of this SMA with $r_{\text{cut}} \sim 6$ nearest neighbors;
- **Positive 1nn vacancy binding energies** as in DFT, and in contrast with EAM#3.



⁴Ghanemet et al., J. Appl. Mech. 57 (1990)

⁵Deb et al., IEEE Trans. Evol. Comput., 6 (2002),

⁶Blank et al., IEEE Access, 8 (2020)

⁷Clouet, Phys. Rev. B, 86 (2012)

⁸Varvenne et al., Acta Mater., 78 (2014)

Conclusions

- Sensitivity analysis of SMA interatomic potentials needs to get rid of **artefacts induced by the cutoff**. More generally speaking, **the robustness of a potential is affected by its roughness**;
- We propose a **sensitivity indices-oriented technique** to refit interatomic potentials;
- An **improved SMA** potential is identified, that can be used to **study small point defects physics in α -Zr**. Extended defects will not be well described, however, as a better description of stacking faults would be necessary;
- Further efforts to obtain a better SMA potential require (i) to treat the cutoff radius explicitly as an adjustable parameter and to optimize it, and (ii) a more global SMA model screening, in order to push the optimization problem up to the SMA expressivity limits.