Modified Stillinger-Weber Interatomic Potentials

Molecular dynamics simulation of vapor deposition of elemental and compound materials

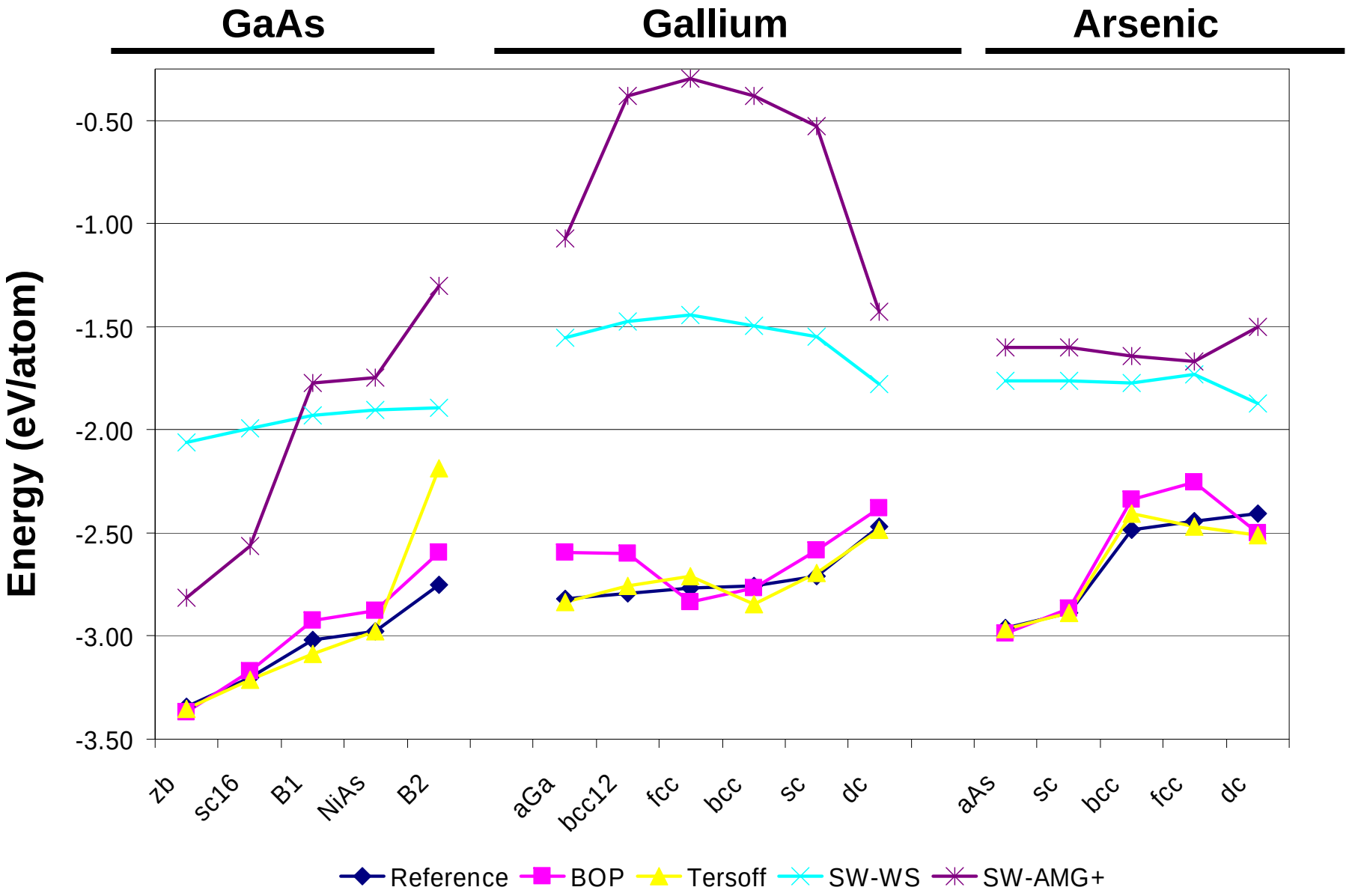
X. W. Zhou

1. X. W. Zhou and H. N. G. Wadley, Computational Materials Science, in press, 2007.
2. X. W. Zhou and H. N. G. Wadley, Computational Materials Science, in press, 2007.

Objectives

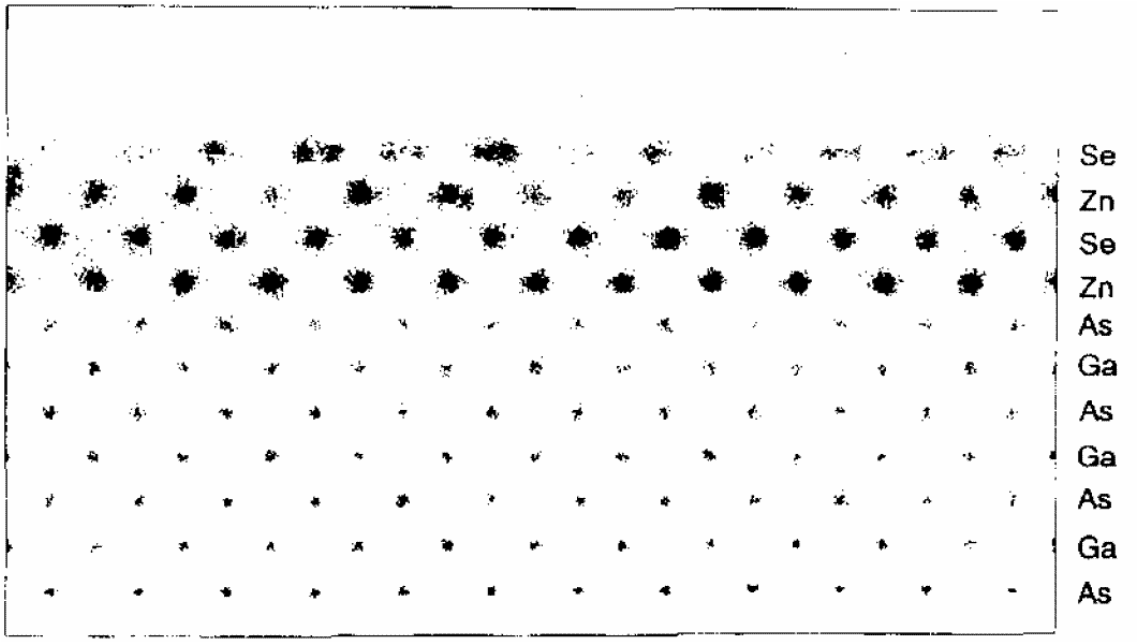
1. Understanding better the Stillinger-Weber potentials.
   - why we decide to use it?
   - where it may not be accurate?
2. Extending the Stillinger-Weber potentials.
   - dc, sc, bcc, and fcc elements.
   - B1, B2, and B3 binary compounds.
3. Simulating growth and oxidation of materials using molecular dynamics methods.
   - case studies: Si, Po, Fe, Ni, Mg, and MgO.

Cohesive Energy



D. A. Murdick, X. W. Zhou, and H. N. G. Wadley, *Phys. Rev. B*, **72**, 205340(2005).

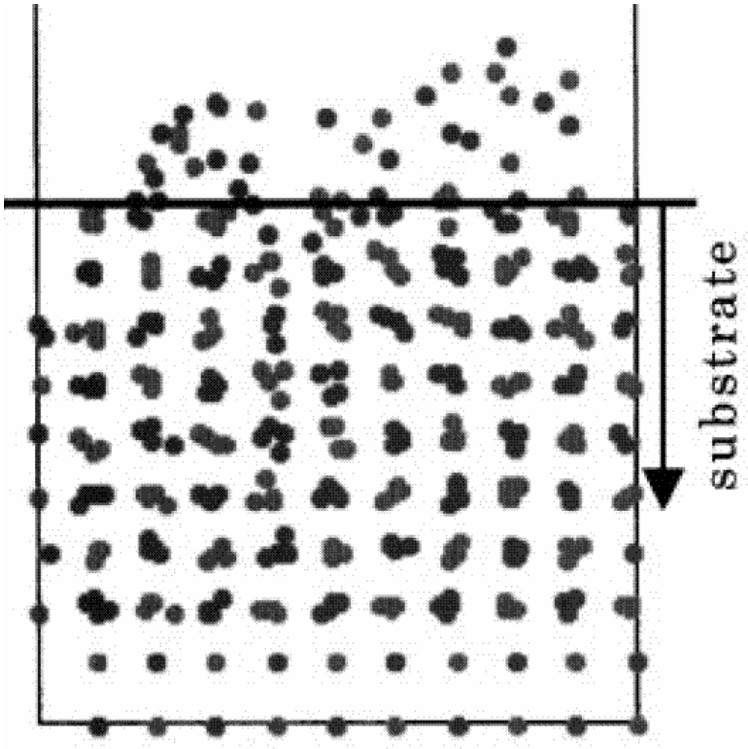
Published Growth Simulation Using SW Potential



Growth of ZnSe on As-terminated (001) GaAs surface. – from Ref. [1].

1. G. H. Grein, J. P. Faurie, V. Bousquet, E. Tournie, R. Benedek, and T. de la Rubia, *J. Cryst. Growth*, *178*, 258(1997).
2. G. H. Gilmer, and C. Roland, *Appl. Phys. Lett.*, **65**, 824(1994).
3. B. Strickland, and C. Roland, *Phys. Rev. B*, **51**, 5061(1995).
4. H. W. Lu, and J. Y. Feng, *Modelling Simul. Mater. Sci. Eng.*, **8**, 621(2000).

Published Growth Simulation Using TF Potential



Growth of InAs on (110) GaAs surface. –from Ref. [1].

All the potentials listed on this slide do not correctly predict the crystalline growth. The Tersoff Si potential, Ref. [2], for instance, predicts the crystalline growth only at temperature above 2200 K.

1. M. Nakamura, H. Fujioka, K. Ono, M. Takeuchi, T. Mitsui, and M. Oshima, *J. Cryst. Growth*, **209**, 232(2000).
2. J. Tersoff, *Phys. Rev. B*, **39**, 5566(1989). – for Si.
3. P. A. Ashu, J. H. Jefferson, A. G. Cullis, W. E. Hagston, and C. R. Whitehouse, *J. Cryst. Growth*, **150**, 176(1995). – for GaAs.
4. R. Smith, *Nucl. Instru. Meth. B*, **67**, 335(1992). – for GaAs.

Formalisms

Stillinger-Weber potential:

$$E_c = \frac{1}{2N}\sum_{i=1}^{N}\sum_{j=i_1}^{i_N}\phi(r_{ij}) + \frac{1}{2N}\sum_{i=1}^{N}\sum_{j=i_1}^{i_N}\left[\left(\sum_{\substack{k=i_1 \\ k\neq j}}^{i_N} u(r_{ik})\cdot g(\cos\theta_{jik})\right)\cdot u(r_{ij})\right]$$

Tersoff potential:

$$E = \frac{1}{2N}\sum_{i=1}^{N}\sum_{j=i_1}^{i_N} V_R(r_{ij}) + \frac{1}{2N}\sum_{i=1}^{N}\sum_{j=i_1}^{i_N}\overline{B_{ij}}\cdot V_A(r_{ij})$$

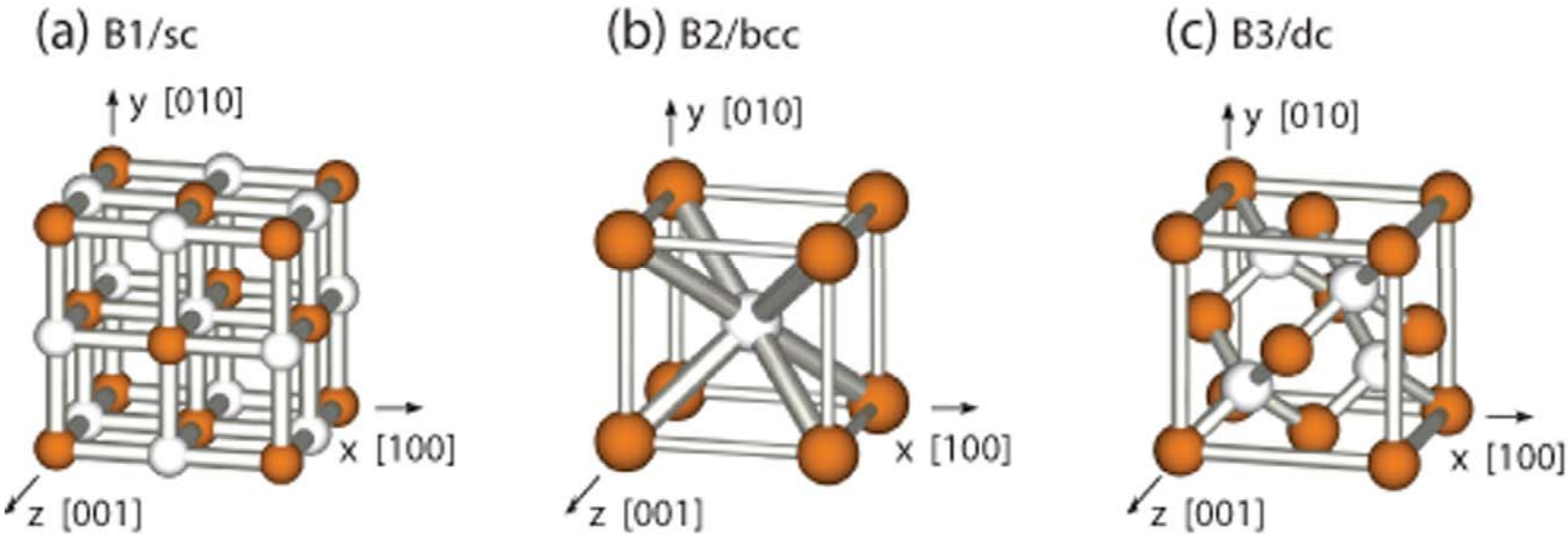
$$\phi(r) = A\cdot S\cdot\left(\frac{\sigma}{r}\right)^4\cdot\exp\left(\frac{\sigma}{r-r_c}\right) - A\cdot\exp\left(\frac{\sigma}{r-r_c}\right)$$

$$g(\cos\theta) = \left(\cos\theta + \frac{1}{3}\right)^2$$

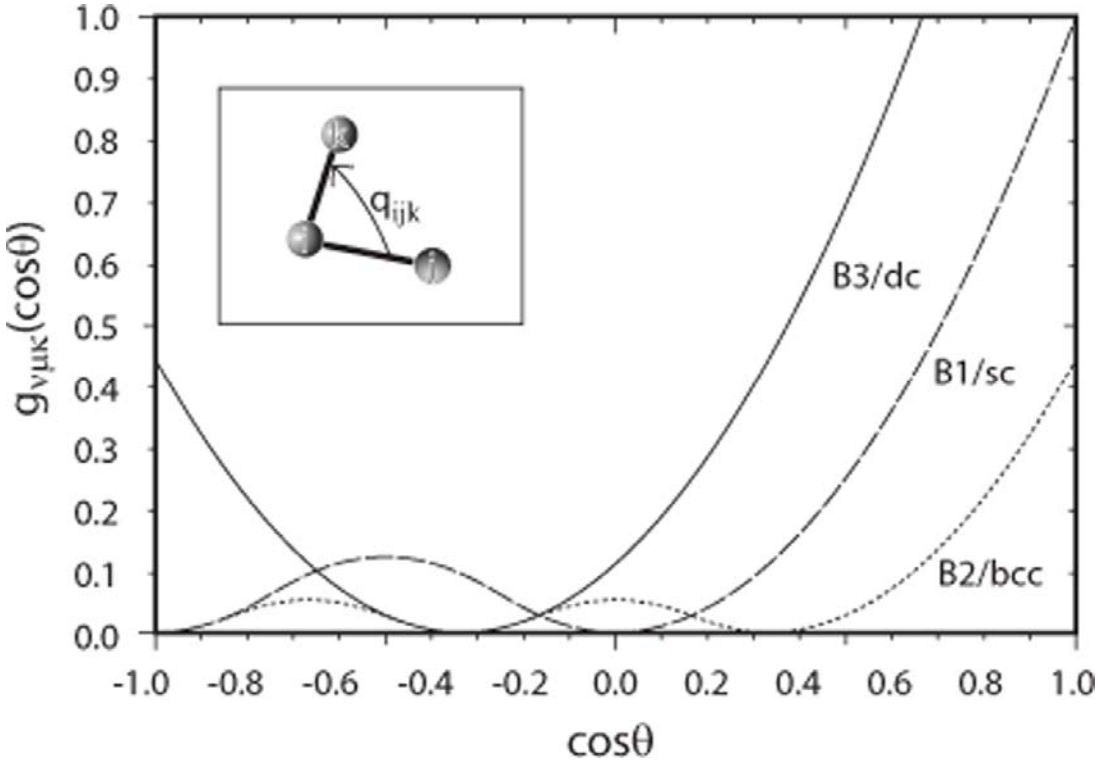
$$u(r) = C\cdot\exp\left(\frac{\gamma}{r-r_c}\right)$$

Keys: Fit f for the equilibrium structure, adjust u to ensure “phase transferability”

Crystal Analysis



Angular Function



$$g_i(\cos\theta) = g_{o,n} + \chi_n \cdot (\cos\theta - \cos\theta_n)^2, \qquad x_{\min,n} \le \cos\theta \le x_{\max,n}, \qquad n = 1,2,\cdots$$

Model Predictions of Properties

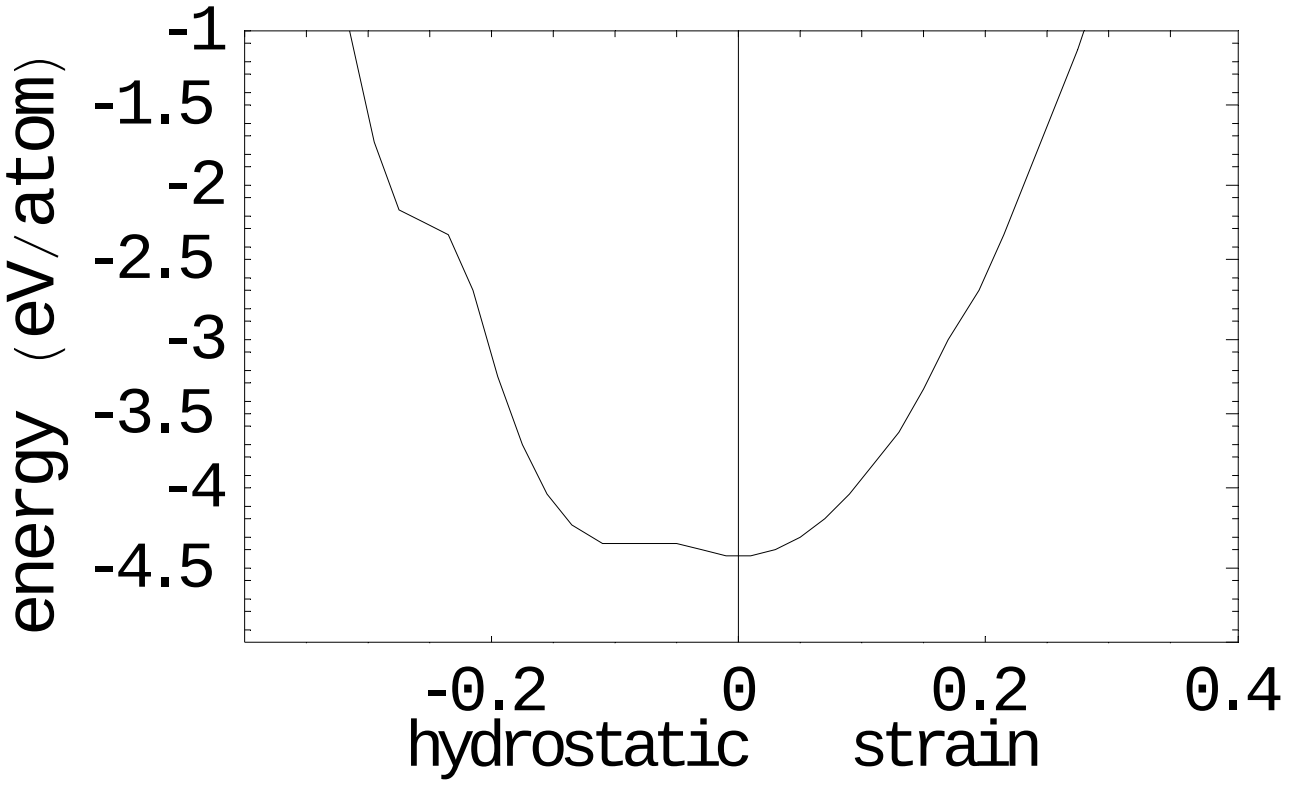
Examples: cohesive energy, lattice constant, bulk modulus, and other elastic constants for four elements

Element	Data type	Cohesive energy, E_c (eV/atom)	Lattice constant, a (Å)	Elastic constants (eV/Å^3)			
				B	C_{11}	C_{12}	C_{44}
Si	Predicted	-4.670	5.431	0.612	1.006	0.415	0.678
	Target	-4.670	5.431	0.612	1.036	0.400	0.498
Po	Predicted	-1.463	3.280	0.352	1.056	0.000	0.266
	Target	-1.463	3.280	0.352	0.706	0.175	0.266
Fe	Predicted	-4.320	2.866	2.108	2.809	1.757	1.757
	Target	-4.320	2.866	1.054	1.413	0.875	0.725
Ni	Predicted	-4.450	3.520	2.820	4.487	1.987	2.225
	Target	-4.450	3.520	1.128	1.541	0.922	0.779

$$\phi(r) = A \cdot S \cdot \left(\frac{\sigma}{r}\right)^4 \cdot \exp\left(\frac{\sigma}{r - r_c}\right) - A \cdot \exp\left(\frac{\sigma}{r - r_c}\right)$$

Problems of Exactly Fitting the Bulk Modulus

Example: fcc Ni



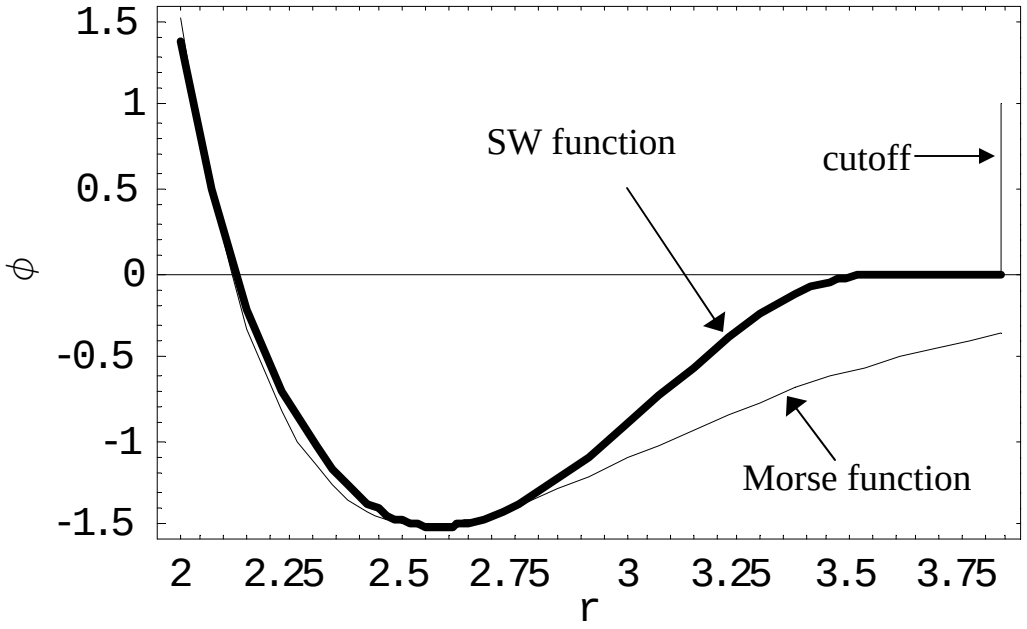
$$\phi(r) = A \cdot S \cdot \left(\frac{\sigma}{r}\right)^4 \cdot \exp\left(\frac{\sigma}{r - r_c}\right) - A \cdot \exp\left(\frac{\sigma}{r - r_c}\right)$$

Smooth function requires S<<A

Comparison SW and Morse Pair Potentials

SW: $$\phi(r) = A \cdot S \cdot \left(\frac{\sigma}{r}\right)^4 \cdot \exp\left(\frac{\sigma}{r - r_c}\right) - A \cdot \exp\left(\frac{\sigma}{r - r_c}\right)$$

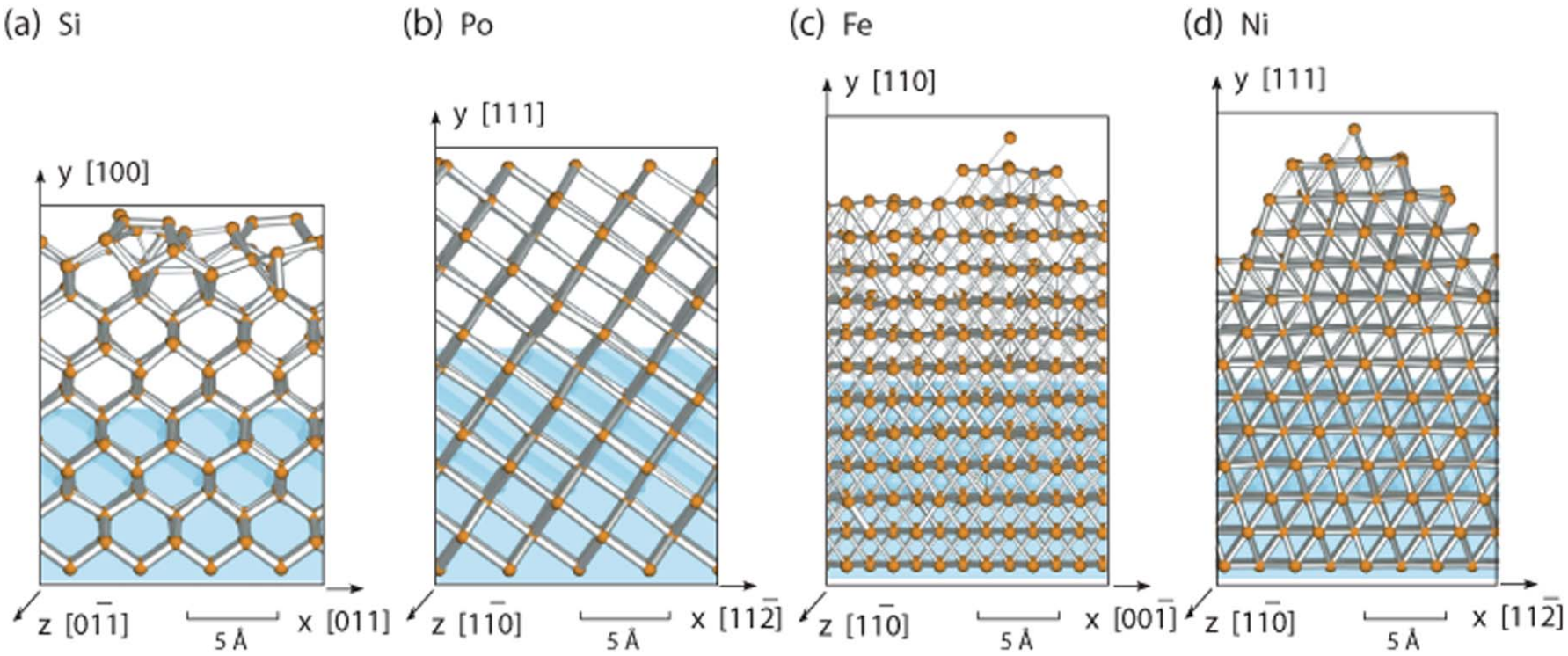
Morse: $$\phi(r) = D_e \cdot \exp\left[-2(r - r_e)\right] - 2D_e \cdot \exp\left[-(r - r_e)\right]$$



Si data from: X. W. Zhou and H. N. G. Wadley, Computational Materials Science, in press, 2006.

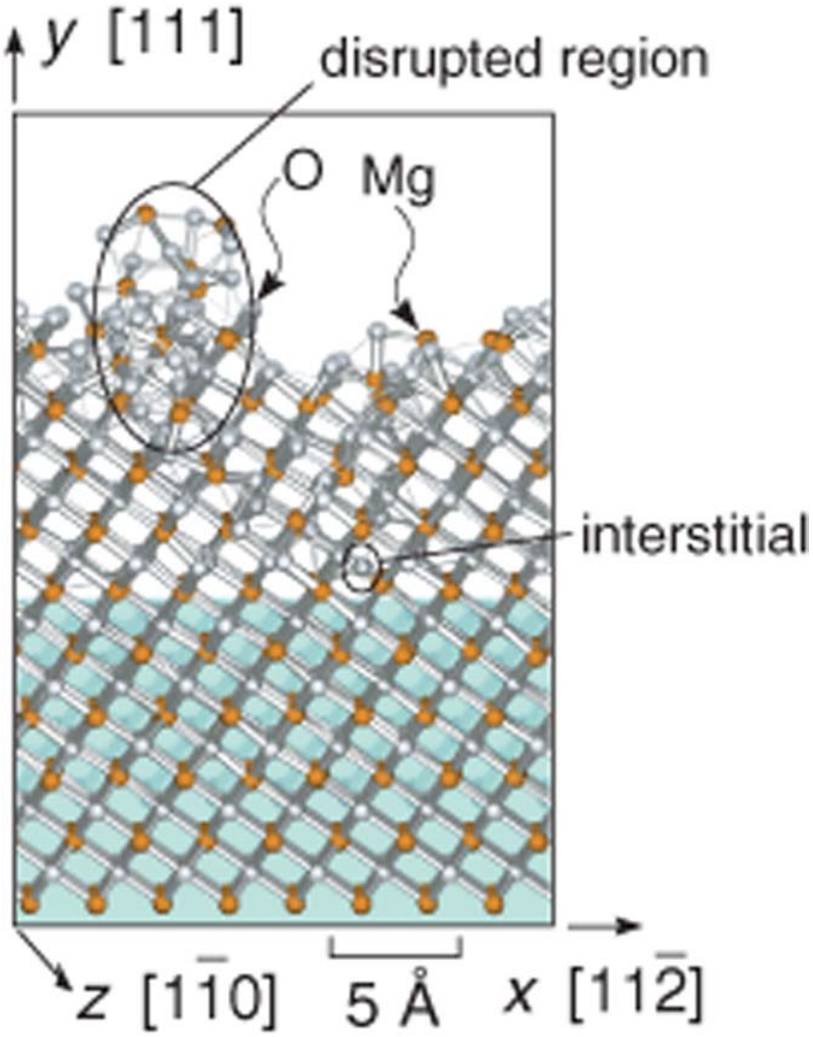
Simulation of Growth: Elemental Systems

Adatoms are randomly injected to the surface at a temperature of 650 K.



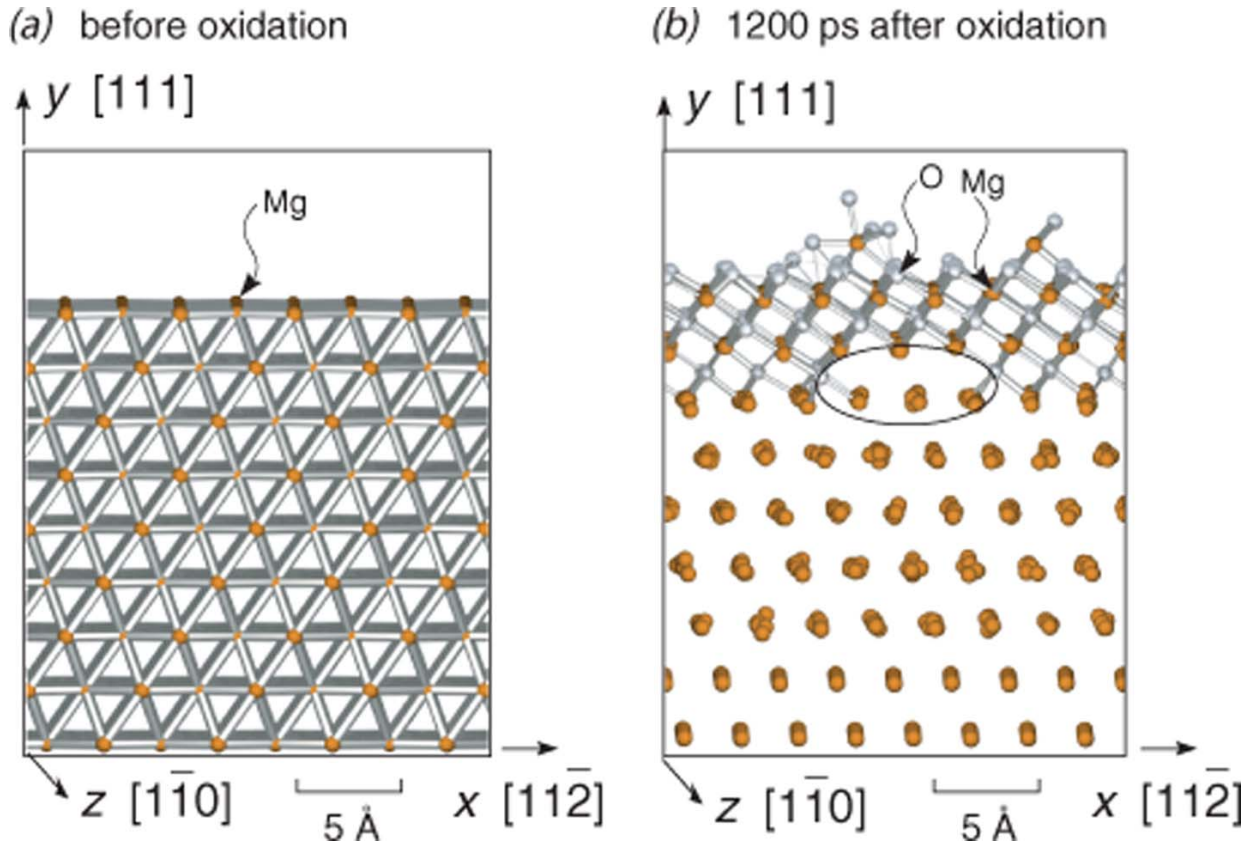
Simulation of MgO Growth

Identical fluxes of Mg and O vapor atoms are randomly injected to the MgO surface at a temperature of 650 K.



Simulation of Growth: Compound Systems

An (111) Mg surface is exposed to oxygen environment at a temperature of 800 K.



Conclusions

1. **Stillinger-Weber potentials only target the equilibrium crystal structure. The energy and crystal geometry of other phases cannot be precisely predicted. As a result, they should not be used in the potential parameterization.**
2. **Extremely easy to parameterize for the equilibrium crystal phase to have the lowest cohesive energy.**
3. **The current pair function forms have nice cutoff, but are not physical and cannot well predict bulk moduli for non- diamond cubic or zinc-blende crystals. This can be easily improved.**
4. **Successfully used to simulate the growth and oxidation of variety materials: Si, Po, Fe, Ni and MgO.**
5. **Stillinger-Weber potentials do not capture energies of molecular gases and therefore cannot reveal the effects of flux ratio of these gases during surface growth.**
6. **Virtual cutoff distance can be reduced to accelerate simulations without affecting the results.**