Exploring atomistic energy landscapes via bifurcation theory, numerical continuation and deflation techniques

Maciej Buze

31 March 2023

Joint work with J. Kermode, L. Shenoy (Warwick Engineering),
T. Hudson (Warwick Mathematics), C. Ortner (UBC)

IPAM Workshop: Increasing the Length, Time, and Accuracy of Materials Modeling Using Exascale Computing
UCLA, 27-31 March 2023

Setup

- A d-dimensional system of N atoms occuppying positions

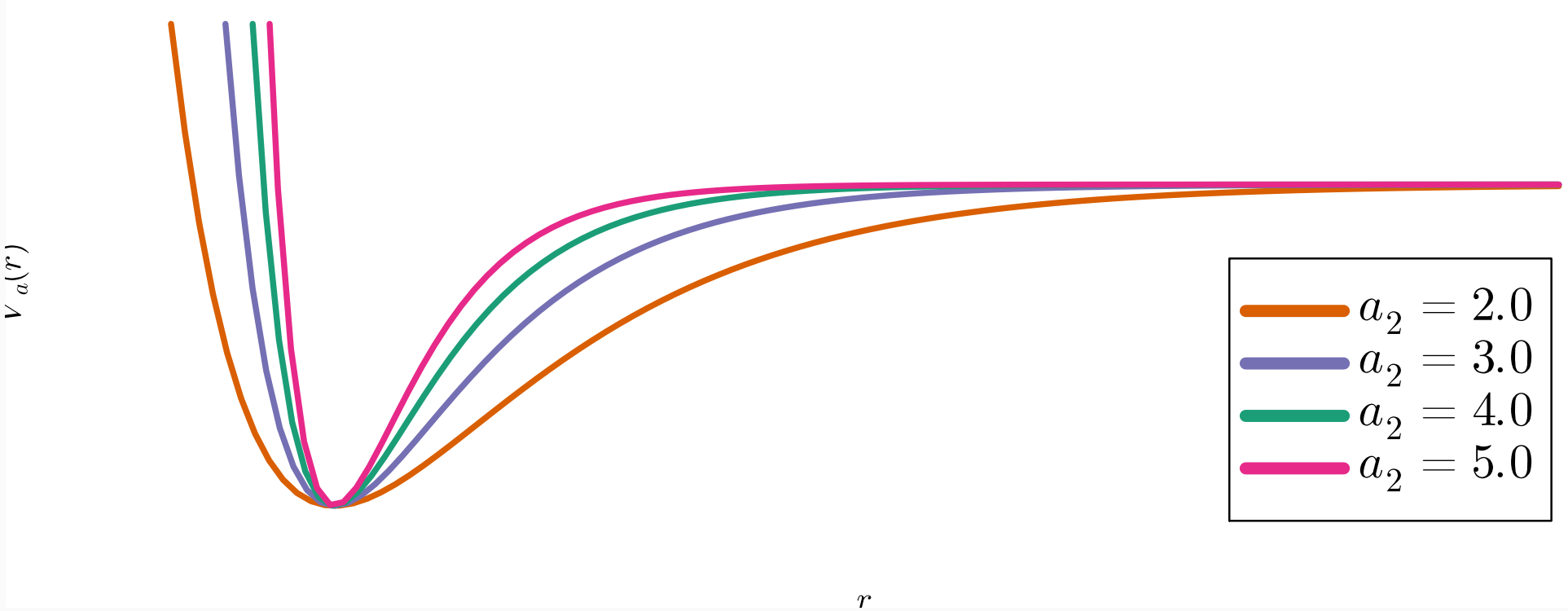
$$\boldsymbol{R} = \{\boldsymbol{r_j}\}_{j=1}^{N} \in (\mathbb{R}^d)^N.$$

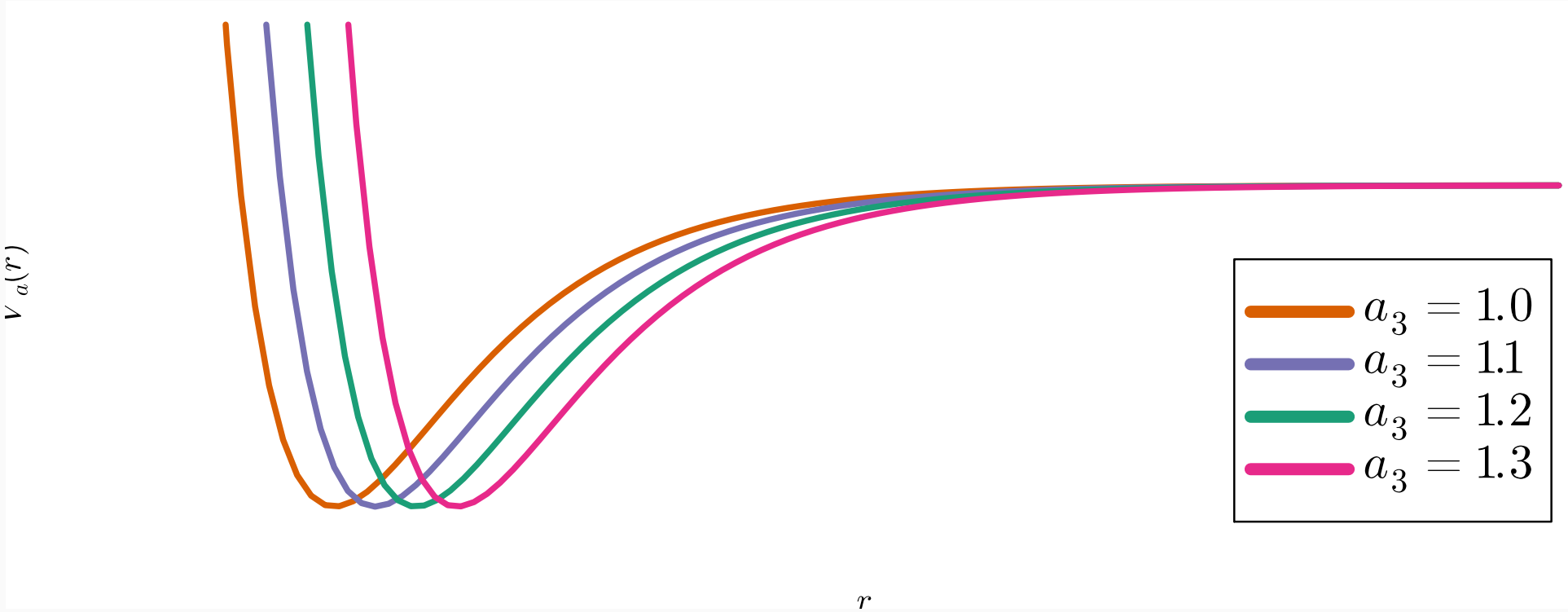
- Atomistic energy $E : (\mathbb{R}^d)^N \to \mathbb{R}$, for instance, given by

$$E(\boldsymbol{R}) = \sum_{k=1}^{K} \frac{1}{k!} \sum_{i_1 \neq \ldots \neq i_k} E_k(\boldsymbol{r_{i_1}}, \ldots, \boldsymbol{r_{i_k}}).$$

- Simple example: $E_1 \equiv 0$, $K = 2$, $E_2(\boldsymbol{r}_1, \boldsymbol{r}_2) = V_{\boldsymbol{a}}(|\boldsymbol{r}_{12}|)$, $\boldsymbol{r}_{12} = \boldsymbol{r}_1 - \boldsymbol{r}_2$ where $\boldsymbol{a} = (a_1, a_2, a_3)$ and

$$V_{\boldsymbol{a}}(r) = a_1 \left(\exp\Big(-2a_2(r - a_3)\Big) - 2\exp\Big(-a_2(r - a_3)\Big) \right).$$


$V_a(r)$
r
$a_2 = 2.0$
$a_2 = 3.0$
$a_2 = 4.0$
$a_2 = 5.0$


$V_a(r)$
r
$a_3 = 1.0$
$a_3 = 1.1$
$a_3 = 1.2$
$a_3 = 1.3$

Numerical continuation and bifurcation theory

- Treat (or modify) E as a function of $\boldsymbol{R}$ and some parameter $\mu \in \mathbb{R}$ ($\mu \in \mathbb{R}^p$ possible), so now $E : (\mathbb{R}^d)^N \times \mathbb{R} \to \mathbb{R}$.
- What can μ be?
  1. $\mu = f(\boldsymbol{a})$, e.g. $f(\boldsymbol{a}) = a_2$ for Morse;
  2. incremental loading at the boundary $\to$ Xiantao Li (Workshop 2), prototyping this approach here with
  3. more "sophisticated" boundary condition approaches $\to$ MB & J.Kermode (Workshop 2).
  4. $E(\boldsymbol{R}, \mu) := \mu E(\boldsymbol{R}) + (1 - \mu) E_{\mathrm{HA}}(\boldsymbol{R}) \to$ Manuel Athenes' talk on Tuesday.
  5. "Helpful biasing force" $\to$ Steve Fitzgerald's talk on Thursday.
  6. ???
- Forces $\boldsymbol{F} : (\mathbb{R}^d)^N \times \mathbb{R} \to (\mathbb{R}^d)^N$ are given by

$$\boldsymbol{F}(\boldsymbol{R}, \mu) = \{-\nabla_{\boldsymbol{r}_i} E(\boldsymbol{R}, \mu)\}_{i=1}^N \equiv \nabla_{\boldsymbol{R}} E(\boldsymbol{R}, \mu).$$

Curves of solutions

- Forces $\boldsymbol{F} : (\mathbb{R}^d)^N \times \mathbb{R} \to (\mathbb{R}^d)^N$ are given by

$$\boldsymbol{F}(\boldsymbol{R}, \mu) = \{-\nabla_{\boldsymbol{r}_i} E(\boldsymbol{R}, \mu)\}_{i=1}^N \equiv \nabla_{\boldsymbol{R}} E(\boldsymbol{R}, \mu).$$

- Set $\mu = \mu^0$ and suppose we have some $\boldsymbol{R}^0$ such that $\boldsymbol{F}(\boldsymbol{R}^0, \mu^0) = 0$ (and that it is regular).
- Implicit Function Theorem implies there exists a curve

$$\big\{(\boldsymbol{R}(\mu), \mu), \quad \mu \in (\mu^0 - \varepsilon, \mu^0 + \varepsilon)\big\}, \quad \boldsymbol{F}(\boldsymbol{R}(\mu), \mu) = 0.$$

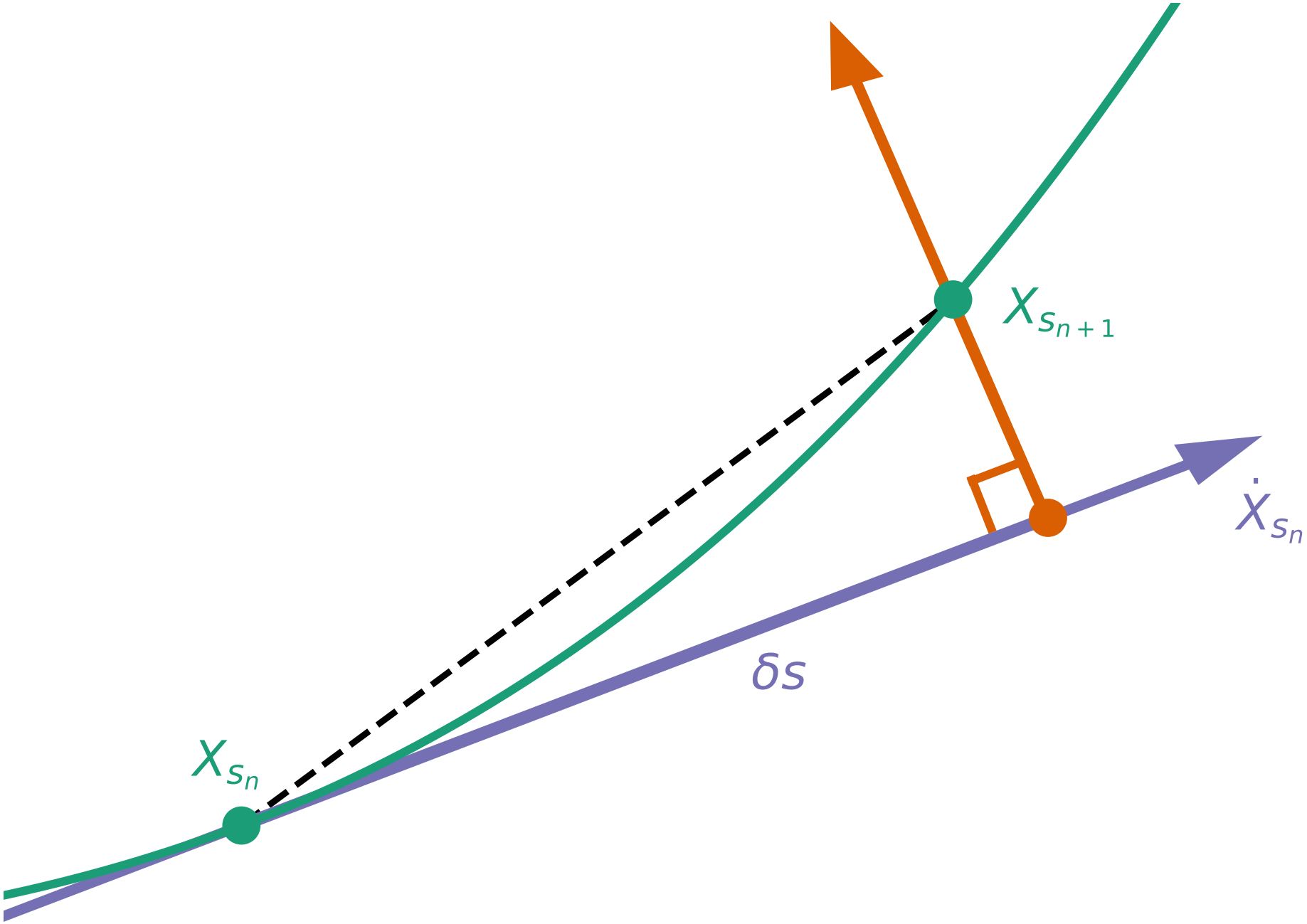
- More generally curves can be parametrised as

$$[0, 1] \ni s \mapsto (\boldsymbol{R}(s), \mu(s)), \quad \boldsymbol{F}(\boldsymbol{R}(s), \mu(s)) = 0.$$

- Constant unit speed parametrisation if $\frac{\mathrm{d}}{\mathrm{d}s}|\boldsymbol{R}(s)|^2 + \frac{\mathrm{d}}{\mathrm{d}s}\mu(s)^2 = 1$.

Numerical continuation

- Assume a curve
$$[0,1] \ni s \mapsto (\boldsymbol{R}(s), \mu(s)), \quad \boldsymbol{F}(\boldsymbol{R}(s), \mu(s)) = 0.$$
exists and that we identified one point on it:
$$X^0 := (R(s^0), \mu(s^0)), \quad \boldsymbol{F}(\boldsymbol{R}(s^0), \mu(s^0)) = 0.$$
- Compute (or approximate) tangent at X^0, ie.
$$\dot{X}^0 := \frac{\mathrm{d}}{\mathrm{d}s} X \Big|_{s=s^0}.$$
- Find the new point on the curve $X^1 = (\boldsymbol{R}(s^1), \mu(s^1))$ by solving
$$\begin{cases} \boldsymbol{F}(\boldsymbol{R}(s^1), \mu(s^1)) & = 0, \\ (X^1 - X^0) \cdot \dot{X}^0 & = ds, \end{cases}$$
where $ds \in \mathbb{R}$ is a parameter we set.
- The extended system can be proven to "see" turning points as regular solutions!


$X_{s_{n+1}}$
$\dot{X}_{s_n}$
δs
X_{s_n}

Application to crack propagation: NCFlex scheme

- In atomistic simulation one often applies K-field boundary condition from continuum elasticity.
- One should combine it with flexible boundary approach*

$$K\,U_{\mathrm{CLE}}(x-\alpha) \approx K\,U_{\mathrm{CLE}}(x) + K\sum_j \alpha_1^j\,U^{(j)}(x).$$

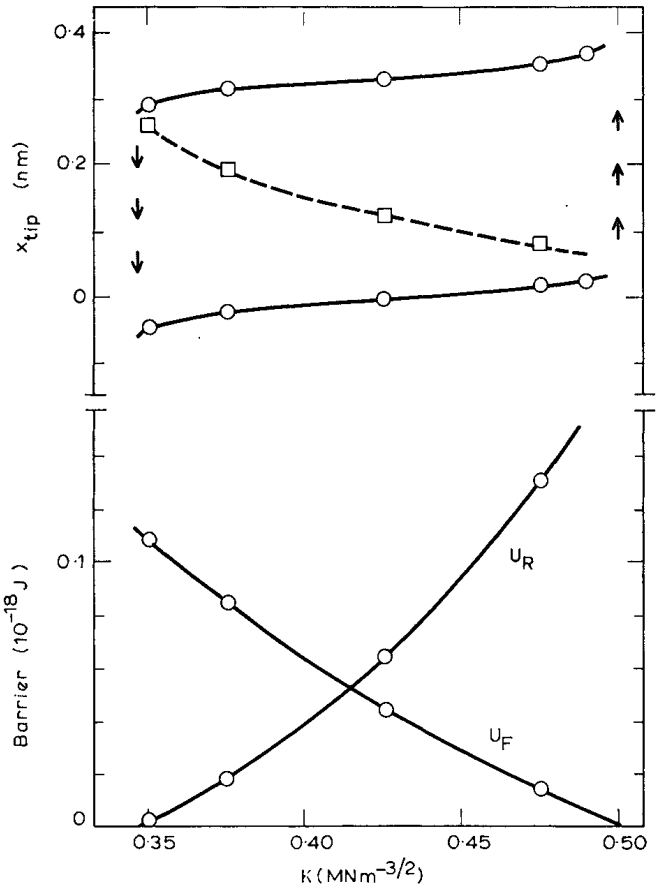
- (We can prove that Sinclair's order expansion is wrong and systematically derive the correct expansion†.)

---

*J.E. Sinclair. The influence of the interatomic force law and of kinks on the propagation of brittle cracks. The Philosophical Magazine: A Journal of Theoretical Experimental and Applied Physics. 1975 Mar 1;31(3):647-71.

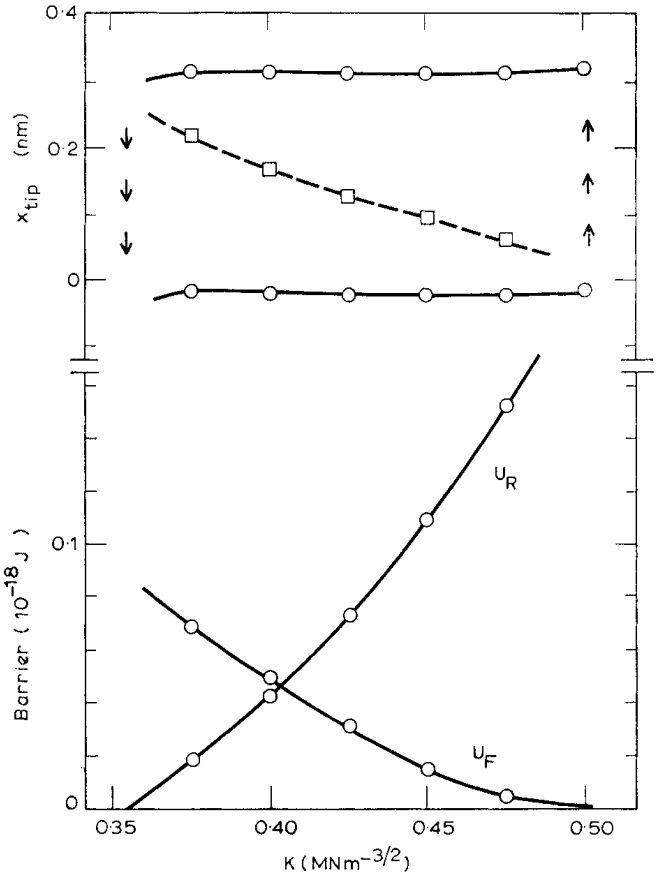
†J. Braun, M.B., in preparation.

Fig. 6



As for fig. 5, but with potential II.

Fig.7



As for fig. 5, but with potential III.

NCFlex algorithm*

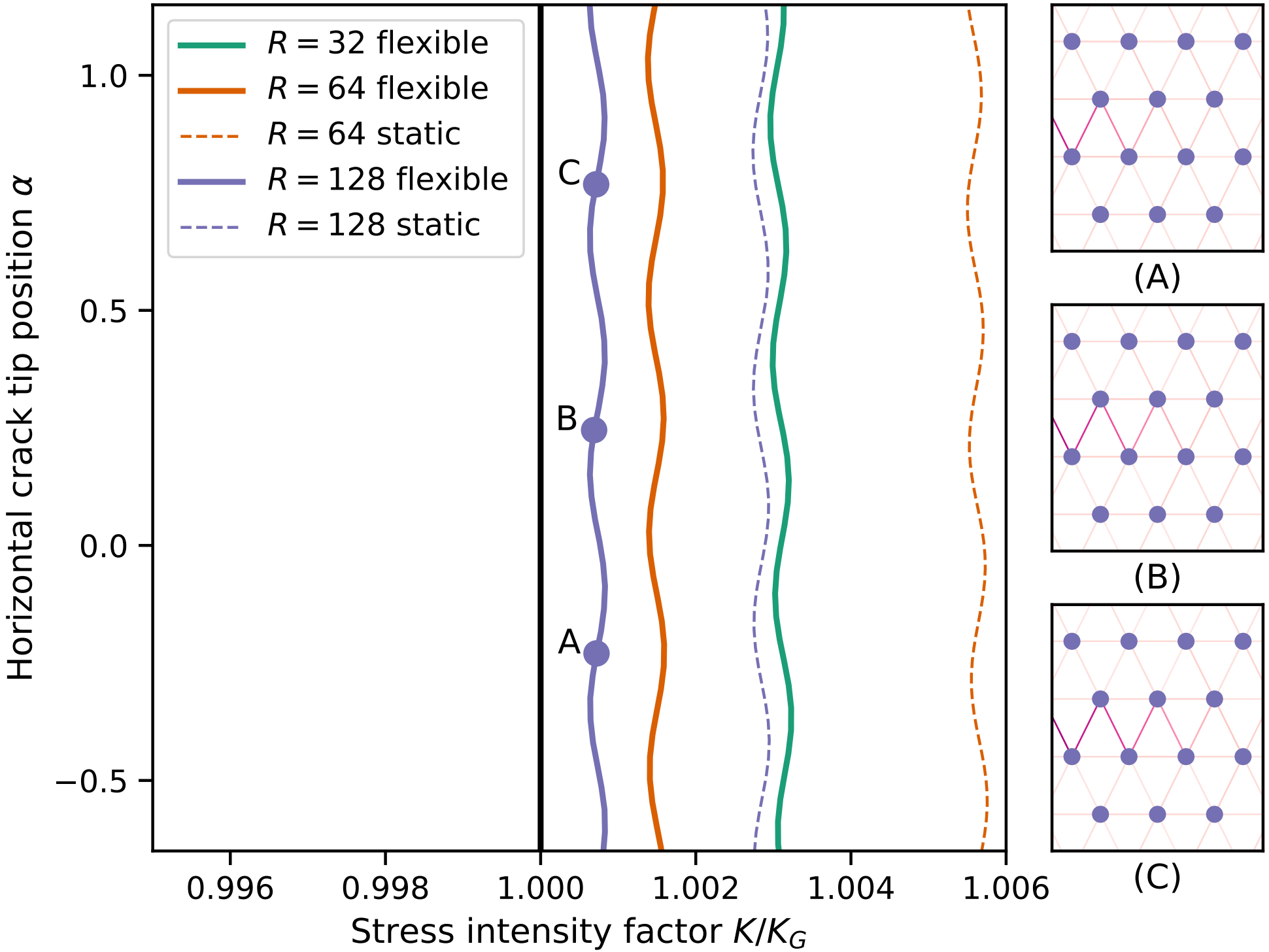
- We use K as a continuation parameter.
- The boundary condition is

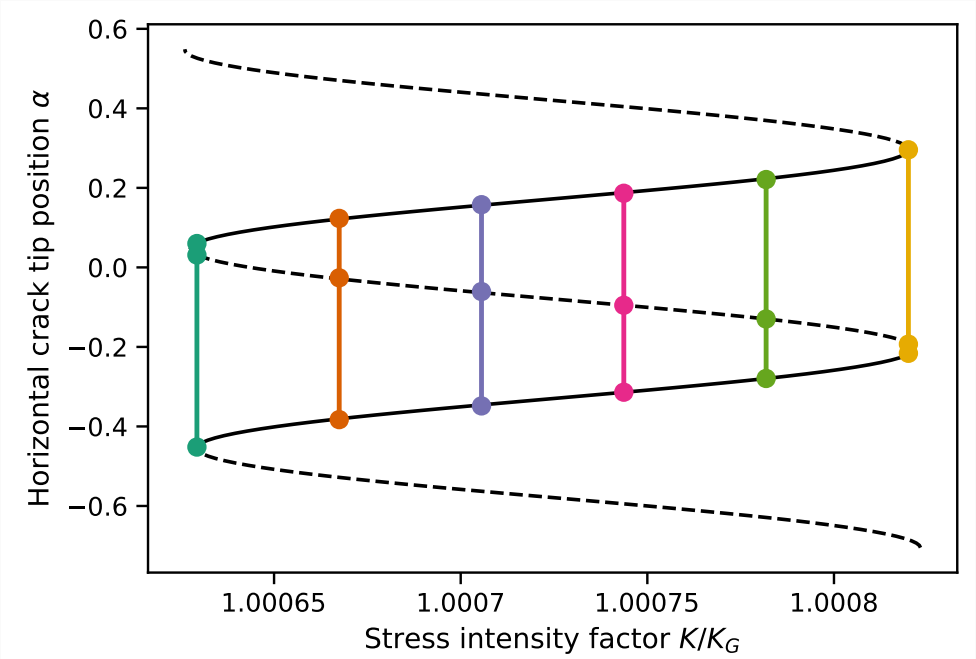
$$K\,U_{\mathrm{CLE}}(x-\alpha).$$

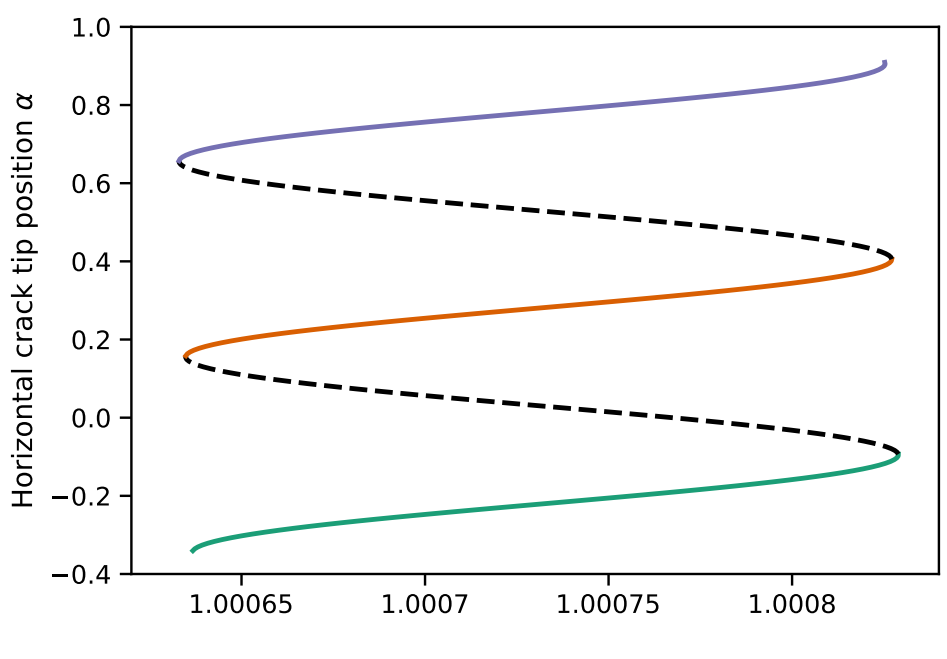
- We apply it first to a toy model Mode III fracture problem: we
  1. analyse finite size effects,
  2. show how bad static boundary conditions are,
  3. derive a simple diagnostic tool for estimating the range of K for which equilibria exist.
- We then apply it to Mode I fracture propagation in silicone using state-of-the-art empirical potentials (Kumagai and Tersoff with screening).
- Ongoing work to use EAM and GAP potentials in BCC Iron.

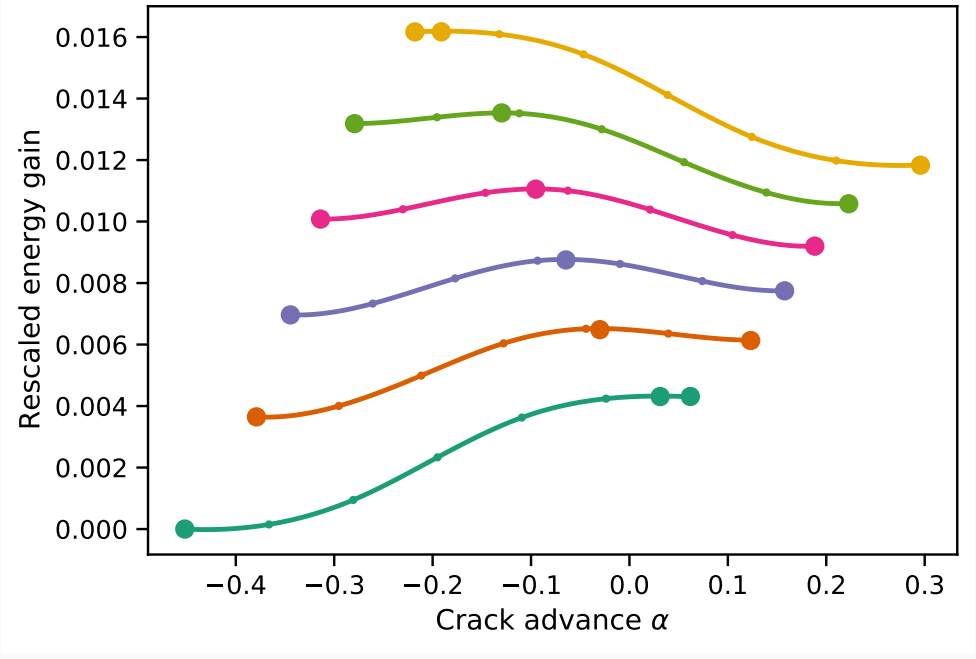
---

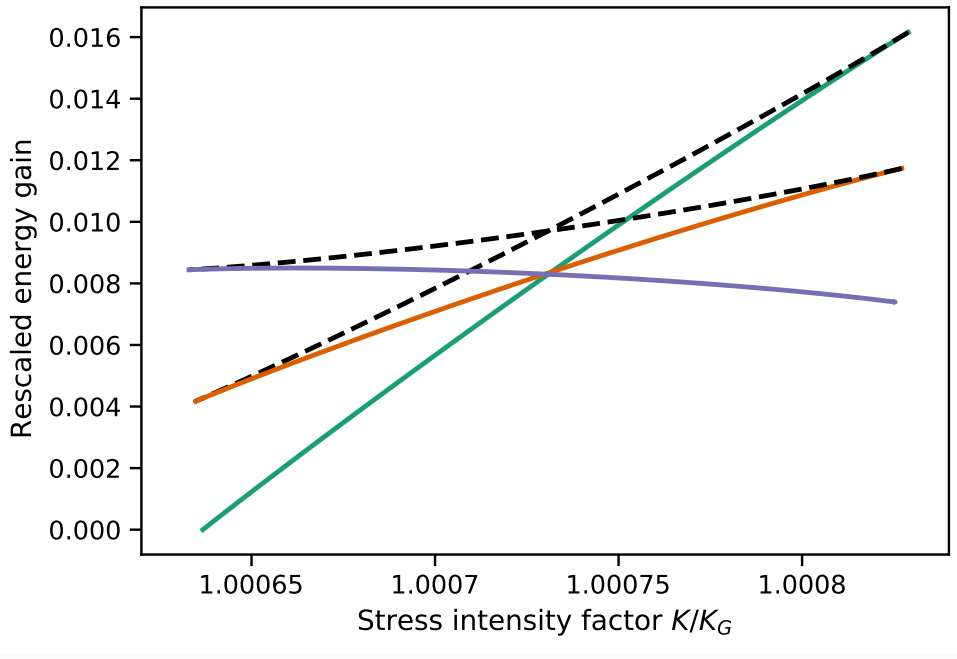
*M. B, J.R. Kermode. Numerical-continuation-enhanced flexible boundary condition scheme applied to mode-i and mode-iii fracture. Physical Review E. 2021 Mar 5;103(3):033002.

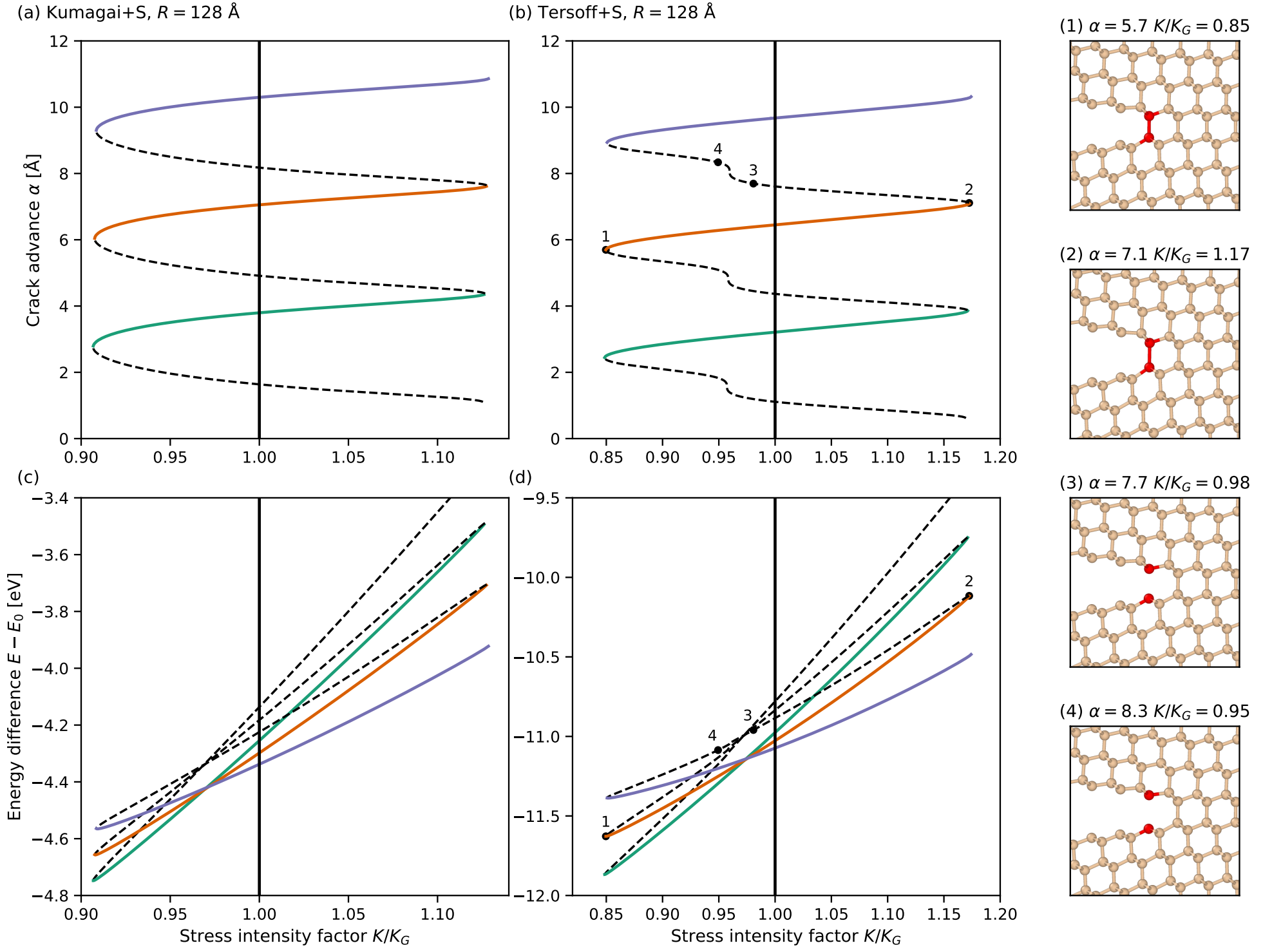

$R = 32$ flexible
$R = 64$ flexible
$R = 64$ static
$R = 128$ flexible
$R = 128$ static
C
B
A
(A)
(B)
(C)
1.0
0.5
0.0
−0.5
0.996
0.998
1.000
1.002
1.004
1.006
Horizontal crack tip position α
Stress intensity factor K/K_G


Horizontal crack tip position α
0.6
0.4
0.2
0.0
−0.2
−0.4
−0.6
1.00065
1.0007
1.00075
1.0008
Stress intensity factor K/K_G


Horizontal crack tip position α
1.0
0.8
0.6
0.4
0.2
0.0
−0.2
−0.4
1.00065
1.0007
1.00075
1.0008


Rescaled energy gain
0.016
0.014
0.012
0.010
0.008
0.006
0.004
0.002
0.000
−0.4
−0.3
−0.2
−0.1
0.0
0.1
0.2
0.3
Crack advance α


Rescaled energy gain
0.016
0.014
0.012
0.010
0.008
0.006
0.004
0.002
0.000
1.00065
1.0007
1.00075
1.0008
Stress intensity factor K/K_G


(a) Kumagai+S, $R = 128$ Å
(b) Tersoff+S, $R = 128$ Å
(c)
(d)
Crack advance α [Å]
Energy difference $E - E_0$ [eV]
Stress intensity factor K/K_G
Stress intensity factor K/K_G
1
2
3
4
(1) $\alpha = 5.7$ $K/K_G = 0.85$
(2) $\alpha = 7.1$ $K/K_G = 1.17$
(3) $\alpha = 7.7$ $K/K_G = 0.98$
(4) $\alpha = 8.3$ $K/K_G = 0.95$

Deflation techniques

- Developed in the context of finding distinct solutions of PDEs by P. Farrell*.
- Original formulation for finding roots of polynomials

$$p(x) = \sum_{k=1}^{N} a_k x^k = \prod_{k=1}^{N} (x - x_k).$$

- If we know $p(x_1) = 0$, then run your root finding algorithm on the *deflated* function

$$q_1(x) = \frac{p(x)}{x - x_1}.$$

- We can iterate: having found x_2 such that $q_1(x_2) = 0$, run your root finding algorithm on

$$q_2(x) = \frac{q_1(x)}{x - x_2} = \frac{p(x)}{\prod_{i=1}^{2}(x - x_i)}.$$

*P.E. Farrell,A. Birkisson,S.W. Funke. Deflation techniques for finding distinct solutions of nonlinear partial differential equations. SIAM Journal on Scientific Computing. 2015;37(4):A2026-45.

Prototype deflation approach in atomistic modelling

- Forces $\boldsymbol{F}: (\mathbb{R}^d)^N \to (\mathbb{R}^d)^N$ are given by

$$\boldsymbol{F}(\boldsymbol{R}) = \{-\nabla_{\boldsymbol{r}_i} E(\boldsymbol{R})\}_{i=1}^N \equiv \nabla_{\boldsymbol{R}} E(\boldsymbol{R}).$$

- Suppose we have identified $\boldsymbol{R}^0$ such that $\boldsymbol{F}(\boldsymbol{R}^0) = 0$.
- We construct a deflated residual

$$\boldsymbol{G}(\boldsymbol{R}) := M_{p,\alpha}(\boldsymbol{R}, \boldsymbol{R}^0)\boldsymbol{F}(\boldsymbol{R}),$$

$$M_{p,\alpha}(\boldsymbol{R}, \boldsymbol{R}^0) := \left(\frac{1}{\|\boldsymbol{R} - \boldsymbol{R}^0\|^p} + \alpha\right)\mathrm{Id}, \quad \alpha \in \mathbb{R}.$$

- Even if the Jacobian of $\boldsymbol{F}$ (the Hessian of the energy) is sparse, the Jacobian of $\boldsymbol{G}$ is dense:

$$\boldsymbol{G}(\boldsymbol{R}) = \eta(\boldsymbol{R})\boldsymbol{F}(\boldsymbol{R}) \implies \delta\boldsymbol{G}(\boldsymbol{R}) = \eta(\boldsymbol{R})\boldsymbol{F}'(\boldsymbol{R}) + \eta'(\boldsymbol{R}) \otimes \boldsymbol{F}(\boldsymbol{R}).$$

- Preconditioning that works well for $\boldsymbol{F}$ will work well for $\boldsymbol{G}$.

Conclusions

- A pretty robust tool for exploring atomistic energy landscapes.
  $\rightarrow$ *"Nonlinear solvers that scale well and always converge (...) and a pony"*
- It works well when there is some "dominating" phenomenon (e.g. fracture in brittle materials).
- One can combine deflation and continuation to sketch out detailed information about the energy landscape.
- Need good continuation parameters.
- Probably need to adjust the deflation operator, e.g.

$$M(\boldsymbol{R}, \boldsymbol{R}^0) = \left(\frac{1}{\|\boldsymbol{R} - \boldsymbol{R}^0\|_{\ell^2(B_r(m))}} + \alpha \right) \mathrm{Id}.$$

- IPAM-faciliated collaboration with Soumendu Bagchi (Los Alamos National Laboratory) to apply this to dislocation nucleation from surface steps.