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A Criterion for Predicting Instabilities in Nanostructures

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Motivation

Many defect nucleation phenomena (dislocation nucleation, crack propagation, grain boundary sliding, etc.) have been characterized as atomic-scale instabilities in which atomic bonds break and reform, shifting the body into a lower-energy configuration.

Various instability criteria have been proposed to model this process: Albers *et al.* (1992), Van Vliet *et al.* (2003), Kitamura *et al*. (2004 a,b), Lu and Zhang (2006a, b), Miller and Rodney (2008), Pacheco and Batra (2008), Delph *et al*. (2009), Delph & Zimmerman (2010).

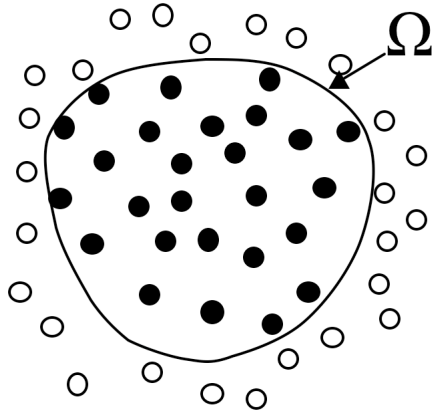
At least for the simple case of pair potentials, these may all be shown to be equivalent at a certain level to a criterion proposed by Wallace (1972).

Wallace's Instability Criterion

Wallace's criterion simply states that a given atomic configuration is stable if all possible infinitesimal atomic displacements from the equilibrium configuration lead to an increase in the system potential energy.

Basic idea: Wallace's method can be efficiently applied to atomistic simulations by examining the behavior of a small (but not too small!) subset of atoms (Miller & Rodney, 2008; Delph *et al*., 2009; Delph & Zimmerman, 2010).

We define a region Ω containing N atoms whose stability we wish to examine. The N atoms are interior to a much larger body, and interact with M atoms in this larger body.



Analysis for pair and multi-body potentials

Assume that atom α, $\alpha = 1,\ldots,N$, originally located at $\mathbf{R}^\alpha$ undergoes an infinitesimal displacement $\mathbf{u}^\alpha$ so that $\mathbf{r}^\alpha = \mathbf{R}^\alpha + \mathbf{u}^\alpha$. All atoms outside of Ω are held fixed.

The potential energy is expressed as: $\Psi = \Psi_2 + \Psi_N$

$$\Psi_2 = \frac{1}{2}\sum_{\alpha=1}^{N_A}\sum_{\gamma=1,\gamma\neq\alpha}^{N_A}\psi_2\left(R^{\alpha\gamma^2}\right) \qquad \Psi_N = \sum_{\delta=1}^{N_A} F\left(\rho^\delta\right)$$

Taylor expansion about the initial equilibrium state gives a quadratic form in the infinitesimal displacements, the linear terms vanishing due to equilibrium.

$$\rho^\delta = \sum_{\varepsilon=1,\varepsilon\neq\delta}^{N_A} f\left(R^{\delta\varepsilon^2}\right)$$

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Taylor expansion of potential energy change

$$\Delta\Psi = \sum_{k=1}^{3N}\sum_{\ell=1}^{3N} A_{k\ell} v_k v_\ell \qquad v_{3(\alpha-1)+i} = u_i^\alpha$$

$$\begin{aligned} A_{3(\alpha-1)+i,3(\alpha-1)+j} = & \sum_{\beta=1,\beta\neq\alpha}^{N} X_{ij}^{\alpha\beta} + \sum_{\gamma=1}^{M} X_{ij}^{\alpha\gamma} \\ & + \sum_{\beta=1,\beta\neq\alpha}^{N} \left[Y_{ij}^{\alpha\beta} + \frac{1}{2}\left(F''(\rho^\alpha) + F''(\rho^\beta)\right) U^{\alpha\beta} U^{\alpha\beta} R_i^{\alpha\beta} R_j^{\alpha\beta} + \right. \\ & \left. \frac{1}{2} F''(\rho^\alpha) \sum_{\eta=1,\eta\neq\alpha,\beta}^{N} U^{\alpha\beta} U^{\alpha\eta} R_i^{\alpha\beta} R_j^{\alpha\eta} + F''(\rho^\alpha) \sum_{\gamma=1}^{M} U^{\alpha\beta} U^{\alpha\gamma} R_i^{\alpha\beta} R_j^{\alpha\gamma} \right] + \\ & \sum_{\gamma=1}^{M} \left(Y_{ij}^{\alpha\gamma} + \frac{1}{2} F''(\rho^\gamma) U^{\alpha\gamma} U^{\alpha\gamma} R_i^{\alpha\gamma} R_j^{\alpha\gamma} \right) + \frac{1}{2} F''(\rho^\alpha) \sum_{\gamma=1}^{M}\sum_{\zeta=1}^{M} U^{\alpha\gamma} U^{\alpha\zeta} R_i^{\alpha\gamma} R_j^{\alpha\zeta} \end{aligned}$$

Taylor expansion - continued

$$
\begin{aligned}
A_{3(\alpha-1)+i,3(\beta-1)+j} \quad = \quad & -X_{ij}^{\alpha\beta} - Y_{ij}^{\alpha\beta} - F''\left(\rho^{\alpha}\right) U^{\alpha\beta} U^{\alpha\beta} R_i^{\alpha\beta} R_j^{\alpha\beta} - \\
& F''\left(\rho^{\alpha}\right) \sum_{\eta=1,\eta\neq\alpha,\beta}^{N} U^{\alpha\beta} U^{\alpha\eta} R_i^{\alpha\eta} R_j^{\alpha\beta} \\
& + \frac{1}{2} \sum_{\eta=1,\eta\neq\alpha,\beta}^{N} F''\left(\rho^{\eta}\right) U^{\alpha\eta} U^{\beta\eta} R_i^{\alpha\eta} R_j^{\beta\eta} + \\
& \sum_{\gamma=1}^{M} \left(-F''\left(\rho^{\alpha}\right) U^{\alpha\beta} U^{\alpha\gamma} R_i^{\alpha\gamma} R_j^{\alpha\beta} + \frac{1}{2} F''\left(\rho^{\gamma}\right) U^{\alpha\gamma} U^{\beta\gamma} R_i^{\alpha\gamma} R_j^{\beta\gamma} \right)
\end{aligned}
$$

$$
X_{ij}^{\alpha\beta} = \frac{1}{2}\left(P^{\alpha\beta}\delta_{ij} + Q^{\alpha\beta} R_i^{\alpha\beta} R_j^{\alpha\beta} \right) \qquad P^{\alpha\beta} = \frac{1}{R^{\alpha\beta}} \frac{\partial \hat{\psi}_2}{\partial R^{\alpha\beta}}
$$

$$
Q^{\alpha\beta} = \frac{1}{R^{\alpha\beta 2}} \left(\frac{\partial^2 \hat{\psi}_2}{\partial R^{\alpha\beta 2}} - \frac{1}{R^{\alpha\beta}} \frac{\partial \hat{\psi}_2}{\partial R^{\alpha\beta}} \right)
$$

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Taylor expansion - continued

$$Y_{ij}^{\alpha\gamma} = \frac{1}{2}\left[\left(F'\left(\rho^{\alpha}\right) + F'\left(\rho^{\gamma}\right)\right)\left(U^{\alpha\gamma}\delta_{ij} + W^{\alpha\gamma}R_i^{\alpha\gamma}R_j^{\alpha\gamma}\right)\right]$$

$$U^{\alpha\beta} = 2f'\left(R^{\alpha\beta^2}\right) \qquad W^{\alpha\beta} = 4f''\left(R^{\alpha\beta^2}\right)$$

Stability of the system of atoms contained within the volume Ω is assured if the quantity $\Delta\Psi$ is positive for any infinitesimal displacement.

This implies that the matrix sym[**A**] must be positive definite, and hence that all real eigenvalues must be positive.

Instability => the appearance of one or more negative real eigenvalues.

$$\left(\mathrm{sym}\left[\mathbf{A}\right] - \theta\mathbf{I}\right)\mathbf{v} = 0$$

$$\Delta\Psi_{\min} = \theta_{\min}$$

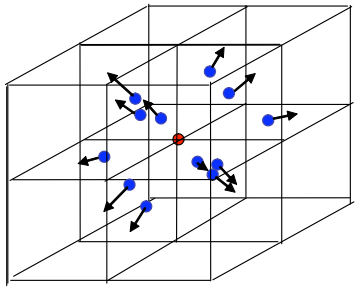
Example: Cavitation in a triaxially stretched FCC crystal

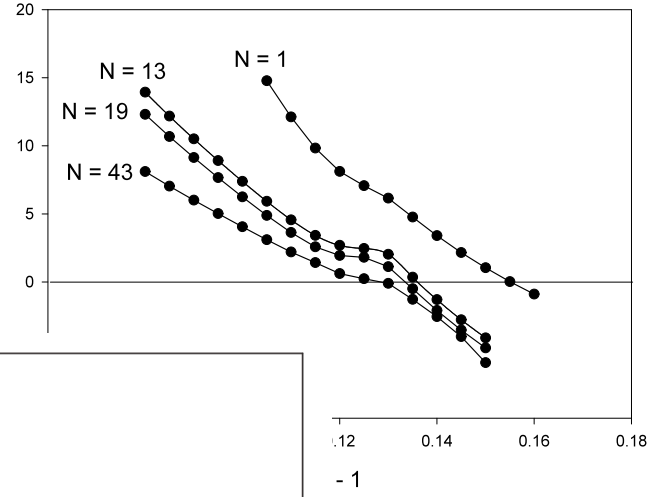
A cavitation type of instability results when a solid is triaxially stretched in tension.

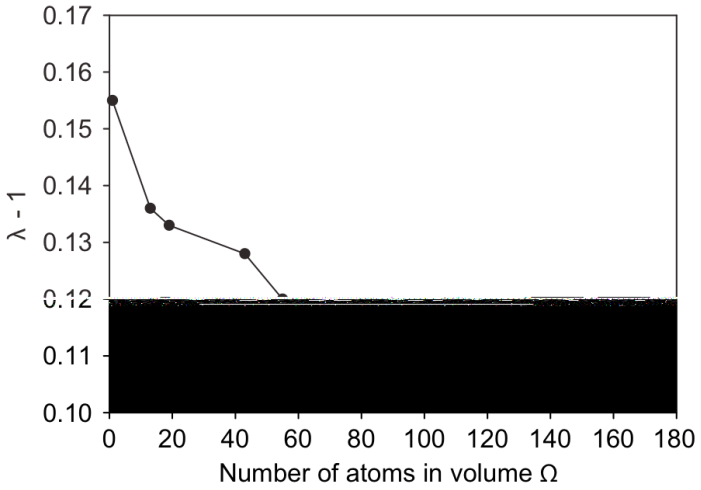
(1) Argon modeled by Lennard-Jones two-body potential by van der Eerden *et al*. (1992)

• Eigenvectors at instability are radially-outward from center atom.

• Stretch ratio at instability is dependent upon the number of atoms considered. The use of small values of *N* leads to inaccurate results.







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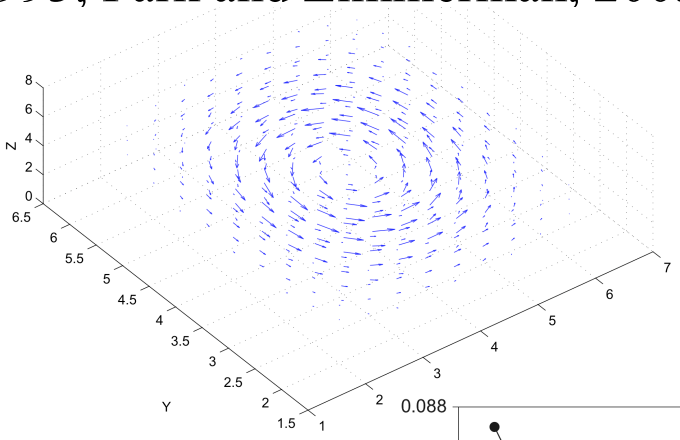
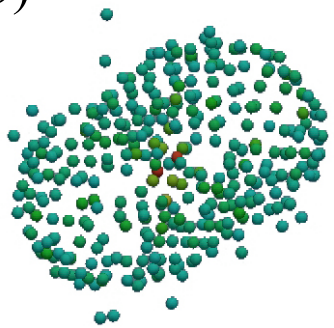
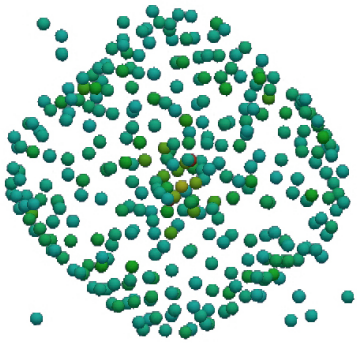
Example: Cavitation - continued

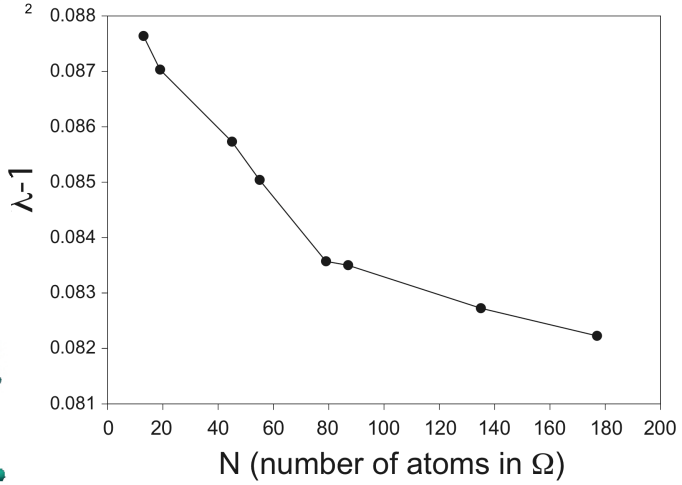
(2) Gold modeled by Embedded Atom Method potential by Foiles (Voter and Chen, 1987; Voter 1993; Park and Zimmerman, 2005)

- A triple root is found for the lowest eigenvalue (Wang *et al*., 1995); oriented cavitation is observed.
- Eigenvectors show isovolumtric motion.
- Independent LAMMPS simulations show an oriented, four-lobe type of cavitation.









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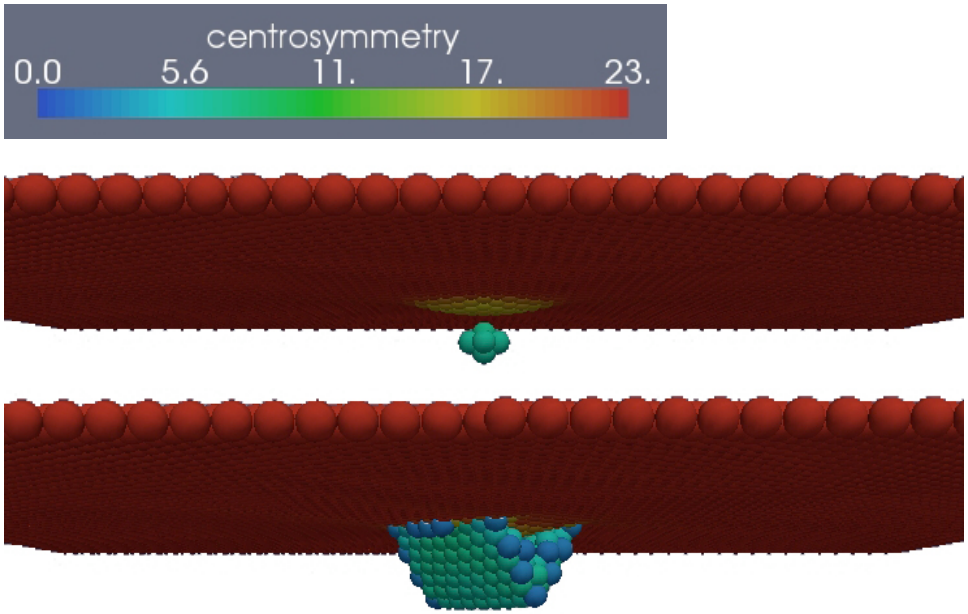
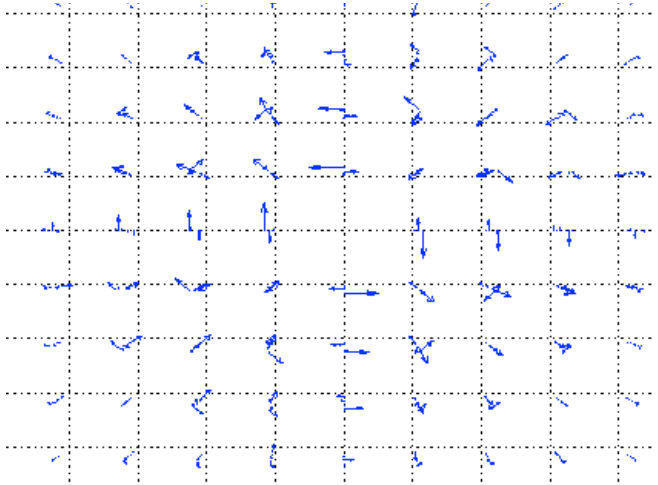
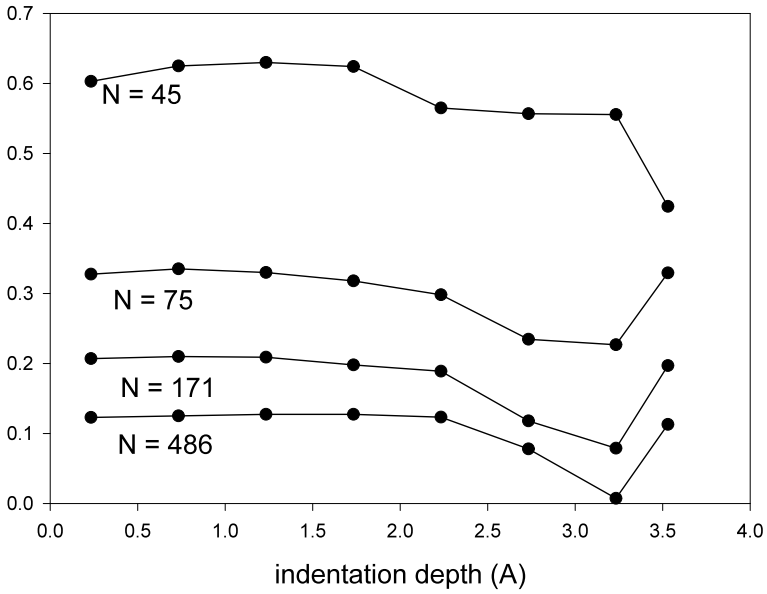
Example: Nanoindentation of gold







- The eigenvectors for lowest eigenvalue indicate a pronounced shearing motion at a depth of 5 atomic layers.
- Dislocation nucleation occurs at this point in LAMMPS simulations.

Concluding Remarks

- We have demonstrated an atomic-scale instability criterion that
  - has a well-defined physical basis and is valid for groups of atoms (*e.g.*, extended defects)
  - makes no reference to continuum quantities
  - leads to accurate predictions of onset and mode of instability
  - is easily computable
  - has been extended to "complicated" inter-atomic potentials
- Future work
  - Develop less computationally expensive ways to generate atomic configurations: Cauchy-Born rule, lattice Green's functions.
  - Use Wallace criterion within multi-scale methods, e.g. Surface Cauchy-Born by Park.
  - Examine stability at finite temperatures: free energy

Miscellanea

- Simulations performed using LAMMPS: http://lammps.sandia.gov

- Thank you to: Jeff Rickman, Joshua Kunz, Harold Park.

- References:
  - Delph *et al*. (*Journal of the Mechanics and Physics of Solids*, 2009)
  - Delph and Zimmerman (*MSMSE*, 2010)

- For more information: jzimmer@sandia.gov