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On numerically predicting the onset and mode of instability in atomistic systems

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ABSTRACT

We present an accurate and efficient method based on the Lanczos algorithm for predicting the onset and mode of instability in atomistic systems. Specifically, we develop a framework that is identically applicable to all flavors of atomistic simulations, including ab-initio calculations. Notably, we do not make any apriori assumptions regarding the nature of the instability or its location. We verify the accuracy of the proposed approach by studying defect nucleation during the nanoindentation of a triangular lattice and hydrostatic tension test of an aluminum crystal. We demonstrate that the computational cost in practical calculations scales linearly with system size, and is accompanied by a small prefactor. Overall, the proposed method is attractive because it enables the stability analysis of atomistic systems at the mesoscale.

1. Introduction

The chemistry of the core, long ranged elastic fields, and discrete nature of the lattice enable crystal defects to have a significant impact on the macroscopic properties of solids [1,2]. This motivates the accurate characterization of defects, which includes developing a fundamental understanding of the mechanisms involved in their nucleation. Since ab-initio calculations are limited by their large computational cost [3–8], atomistic simulations based on empirical potentials have been the preferred choice for studying the nucleation of defects [9–17]. However, these studies focus on the observation rather than prediction of defect nucleation. In particular, the nature of the instabilities—characterized by a sudden rearrangement of atoms resulting in either a structural transformation, phase transition or nucleation of a defect—which give rise to crystal defects is relatively less well understood. Though the present work is targeted toward the atomistic scale, defect nucleation related phenomena can also be studied at higher length scales [18].

In view of the above discussion, there have been previous efforts to predict the onset and mode of instability in atomistic systems [19–22]. However, these approaches employ a complete eigendecomposition of the force constants matrix, i.e., Hessian matrix. Since the computational and memory costs of such methods scale cubically and quadratically with matrix size, respectively, the size of systems that can be studied is severely limited. To overcome this bottleneck in the case of defect nucleation, it has been proposed that the stability analysis be restricted to a suitably chosen subset of atoms [23–25]. However, this strategy assumes the instability mode to be localized, and requires apriori knowledge of the center and size of the localization region to be efficient. Another previously developed technique associates defect nucleation with the loss of positive definiteness of the atomic scale acoustic tensor [26–28]. This requires redefining continuum level quantities at the atomic scale, which can be challenging [29,30]. Furthermore, the instability mode cannot be calculated, and the analysis needs to be performed for every atom, which can make it inefficient.

In this work, we develop a framework based on the Lanczos method for accurately and efficiently predicting the onset and mode of instability in atomistic systems. Notably, the approach scales linearly with respect to the number of atoms in terms of computational cost as well as memory, and possesses a small prefactor, making it a viable choice for large systems. In addition, it does not require any apriori knowledge of the nature or location of the instability, making it widely applicable. We verify the proposed approach by studying defect nucleation during the nanoindentation of a triangular lattice and hydrostatic tension test of an aluminum crystal.

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2. Instability criterion

Consider a system of N atoms in a $S \in \{1,2,3\}$ dimensional space. Let α denote the loading parameter and $\mathbf{r} = \begin{bmatrix} \mathbf{r}_1 & \mathbf{r}_2 & \dots & \mathbf{r}_N \end{bmatrix}^{\mathrm{T}} \in \mathbb{R}^{SN\times 1}$ represent the atomic configuration. On performing a second order expansion of the system's energy $W(\alpha, \mathbf{r})$ about some configuration $\mathbf{r}^*$ (for fixed α), we arrive at

$$W(\alpha,\mathbf{r}) \approx W(\alpha,\mathbf{r}^*) + \mathbf{f}(\alpha,\mathbf{r}^*)^{\mathrm{T}}(\mathbf{r}-\mathbf{r}^*) + \frac{1}{2}(\mathbf{r}-\mathbf{r}^*)^{\mathrm{T}}\mathbf{H}(\alpha,\mathbf{r}^*)(\mathbf{r}-\mathbf{r}^*), \tag{1}$$

where

$$\mathbf{f}(\alpha,\mathbf{r}) = \begin{bmatrix} \dfrac{\partial W}{\partial \mathbf{r}_1} \\ \dfrac{\partial W}{\partial \mathbf{r}_2} \\ \vdots \\ \dfrac{\partial W}{\partial \mathbf{r}_N} \end{bmatrix} \in \mathbb{R}^{SN\times 1} \tag{2}$$

represents the force on the atoms and

$$\mathbf{H}(\alpha,\mathbf{r}) = \begin{bmatrix} \dfrac{\partial^2 W}{\partial \mathbf{r}_1^2} & \dfrac{\partial^2 W}{\partial \mathbf{r}_1\mathbf{r}_2} & \cdots & \dfrac{\partial^2 W}{\partial \mathbf{r}_1\mathbf{r}_N} \\ \dfrac{\partial^2 W}{\partial \mathbf{r}_2\mathbf{r}_1} & \dfrac{\partial^2 W}{\partial \mathbf{r}_2^2} & \cdots & \dfrac{\partial^2 W}{\partial \mathbf{r}_2\mathbf{r}_N} \\ \vdots & \vdots & \ddots & \vdots \\ \dfrac{\partial^2 W}{\partial \mathbf{r}_N\mathbf{r}_1} & \dfrac{\partial^2 W}{\partial \mathbf{r}_N\mathbf{r}_2} & \cdots & \dfrac{\partial^2 W}{\partial \mathbf{r}_N^2} \end{bmatrix} \in \mathbb{R}^{SN\times SN} \tag{3}$$

denotes the Hessian, also referred to as the force constants matrix.

We are interested in predicting the onset and mode of instability as the loading parameter α is varied. To this end, we now define the instability criterion for some given α. Let $\mathbf{r}_\alpha^*$ represent the corresponding equilibrium atomic configuraton, i.e., $\mathbf{f}(\alpha,\mathbf{r}_\alpha^*) = 0$. The stability of the atomistic system can then be determined by the sign of the stability parameter

$$\lambda_\alpha^* = \min_{\substack{\mathbf{v} \\ \|\mathbf{v}\|^2 = 1}} \mathbf{v}^{\mathrm{T}}\mathbf{H}_\alpha^*\mathbf{v}, \tag{4}$$

where $\|\mathbf{v}\|$ represents the l_2 norm of $\mathbf{v}$, and $\mathbf{H}_\alpha^* = \mathbf{H}(\alpha,\mathbf{r}_\alpha^*)$. In particular, the system is unstable if

$$\lambda_\alpha^* < 0, \tag{5}$$

which indicates that the configuration $\mathbf{r}_\alpha^*$ does not correspond to a minimum in the energy landscape, i.e., $W(\alpha,\mathbf{r})$ is not convex about $\mathbf{r}_\alpha^*$. The nature of the mode $\mathbf{v}_\alpha^*$—the minimizer of the variational problem in Eq. (4)—provides information about the type of instability, and can be used to ascertain whether it corresponds to the nucleation of a defect.

It is worth noting that in the above discussion, we do not account for additional constraints that may be prescribed on the system. For example, it is common to hold the positions of some of the atoms fixed in practical calculations. In such a situation, the components of $\mathbf{v}$ corresponding to the fixed atoms need to be enforced to be zero while solving the variational problem in Eq. (4). Equivalently, the stability analysis can be restricted to the movable atoms by eliminating the rows and columns of $\mathbf{H}_\alpha^*$ that correspond to the fixed atoms.

3. Lanczos framework for stability analysis

In order to check the stability of the system at any given loading parameter α, the variational problem in Eq. (4) needs to be solved. The corresponding Euler–Lagrange equation is

$$\mathbf{H}_\alpha^*\mathbf{v} = \lambda\mathbf{v}, \tag{6}$$

where λ is the Lagrange multiplier used to enforce the constraint. It follows that the lowest eigenvalue and corresponding eigenvector of $\mathbf{H}_\alpha^*$ coincide with the stability parameter λ_α^* and mode $\mathbf{v}_\alpha^*$, respectively. Instead of solving for λ_α^* and $\mathbf{v}_\alpha^*$ alone, it is common to eigendecompose $\mathbf{H}_\alpha^*$ [19–22], i.e., compute all its eigenvalues and eigenvectors. However, the computational and memory costs of such a procedure scale as $\mathcal{O}(N^3)$ and $\mathcal{O}(N^2)$ with respect to the number of atoms, respectively, which severely restricts the size of systems that can be studied. In order to overcome this limitation, it has been proposed that the stability analysis be restricted to a suitably chosen subset of atoms [23–25]. However, such techniques assume $\mathbf{v}_\alpha^*$ to be localized, and even then they need apriori knowledge of the location and size of the localization region to be efficient. In addition, they require the original ordering of the eigenvalues to remain unaffected by the localization. Such assumptions/approximations can lead to inaccurate results, as demonstrated by the results in Section 4.

In view of the above discussion, we propose calculating λ_α^* and $\mathbf{v}_\alpha^*$ using the implicitly restarted [31] version of the Lanczos method [32] outlined in Algorithm 1. In the Lanczos method, a three term recurrence relation is utilized to generate an orthogonal basis $\mathbf{v}_k$ in which $\mathbf{H}_\alpha^*$ is a tridiagonal matrix:

$$\mathbf{J}_k = \begin{bmatrix} a_1 & b_1 & & & \\ b_1 & a_2 & b_2 & & \\ & \ddots & \ddots & \ddots & \\ & & b_{k-2} & a_{k-1} & b_{k-1} \\ & & & b_{k-1} & a_k \end{bmatrix} \in \mathbb{R}^{k\times k}, \tag{7}$$

where a_k and b_k are scalars calculated during the iteration. The eigenvalues of $\mathbf{J}_k$ approximate those of $\mathbf{H}_\alpha^*$, with the algebraically smallest and largest ones being the first to converge. Thereafter, the transformation matrix $\mathbf{V}$ consisting of the Lanczos vectors $\mathbf{v}_k$ as columns is used to determine the eigenmode $\mathbf{v}_\alpha^*$. In this work, rather than explicitly form the matrix $\mathbf{H}_\alpha^*$, we utilize the finite-difference approximation for the product of $\mathbf{H}_\alpha^*$ with any vector $\mathbf{w}$:

$$\mathbf{H}^*\mathbf{w} \approx \frac{\mathbf{f}(\alpha,\mathbf{r}_\alpha^* + \varepsilon\mathbf{w}) - \mathbf{f}(\alpha,\mathbf{r}_\alpha^*)}{\varepsilon}, \tag{8}$$

where ε is an appropriately small parameter. In doing so, each matrix-vector product is replaced with one force evaluation, since $\mathbf{f}(\alpha,\mathbf{r}_\alpha^*)$ is readily available. Such a strategy is attractive because of the substantial reduction in the computational cost and memory storage requirements, particularly when the interactions are relatively long-ranged. In addition, since the second order derivatives of the energies are not required, the exact same implementation can be interfaced with any flavor of atomistic simulation, including those where the second derivatives are hard/expensive to evaluate, e.g. electronic structure calculations.

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