LOW-DIMENSIONAL PROJECTIONS FOR VISUALIZING ENERGY LANDSCAPES OF ATOMIC SYSTEMS

Vaibhav Bihani
Indian Institute of Technology Delhi
Hauz-Khas, Delhi, India
vaibhav.bihani525@gmail.com

Srikanth Sastry
Jawaharlal Nehru Centre for Advanced Scientific Research
Bengaluru,Karnataka, India
sastry@jncasr.ac.in

Sayan Ranu
Indian Institute of Technology Delhi
Hauz-Khas, Delhi, India
sayanranu@iitd.ac.in

N.M. Anoop Krishnan
Indian Institute of Technology Delhi
Hauz-Khas, Delhi, India
krishnan@iitd.ac.in

ABSTRACT

Understanding the energy landscape is key to discovering materials with targeted properties since the landscape encapsulates system's complete thermodynamic and kinetic behavior, including its non-equilibrium properties, such as relaxation and metastable phases. However, the curse of dimensionality prohibits one from effectively visualizing the energy landscape. Here, we propose a method to visualize the complex energy landscape of atomic systems. We demonstrate that the proposed low-dimensional projection aligns well with the curvatures of the actual landscape, validated through Hessian analysis. Further, we show that we can gain interesting insights into the behavior of different gradient-based and machine-learned optimizers using the proposed visualization approach. We hope that the study will enhance the understanding of the energy landscapes, and contribute to a fundamental understanding of the physics of materials and accelerate their discovery.

1 INTRODUCTION

Exploration of the energy landscapes of systems is a crucial method for investigating materials' fundamental thermodynamic and kinetic behavior (Wales (2018); Frauenfelder et al. (1991); Mauro et al. (2009); Wales & Doye (1997); Wales (2003)) and plays a vital role in understanding and predicting the behavior of materials. The energy landscape of atomic structures can often be characterized as rough, containing numerous local minima, barriers, and non-convex regions (Wales (2003)). Navigating this complex landscape poses a significant challenge in efficiently exploring the search space to identify low-energy configurations. Several optimization algorithms have been developed or adapted to address this challenge in recent years, incorporating strategies such as population-based metaheuristics, machine-learning techniques, and hybrid approaches (Mannan et al. (2024)). However, the mechanisms involved in the optimization are not well understood. Visualizing the optimization trajectory on energy landscape can give insights into the behavior of different optimizers and enable development of better optimization algorithms.

This research paper aims to propose a visualization framework for visualizing the energy landscape of atomic structures. Further we validate the visualised landscape using hessian analysis, and show that our visualisation captures the curvatures present in actual landscape.

The main objectives of this research paper are as follows:

- Develop a general visualisation framework for low-dimensional visualisation of energy landscape of atomic systems.
- Validate the visualised landscape using Hessian based curvature analysis.
- Study the behavior of different optimizers on the visualised landscape.

By addressing these objectives, we aim to advance the field modeling and simulation of complex materials by providing a tool for visualizing the complex energy landscape. The results and insights

presented in this paper will serve as a valuable resource for researchers and practitioners working in the field of materials discovery.

2 PROBLEM FORMULATION

The configuration $\theta(\mathbf{x_1}, \mathbf{x_2}, ...\mathbf{x_N})$ of an atomic system is given by the positions of all the atoms in the system $(\mathbf{x_1}, \mathbf{x_2}, \ldots, \mathbf{x}_N)$ and their types ω_i. Each $\mathbf{x_i}$ represents the position of the i^{th} atom in a d-dimensional space, where d is typically 2 or 3. The potential energy U of an N-atom structure is a function of θ. Specifically, the energy of a system can be written as the summation of one-body $U(r_i)$, two-body $U(r_i, r_j)$, three-body $U(r_i, r_j, r_k)$, up to N-body interaction terms. However, the exact computation of this energy is highly challenging and involves expensive quantum mechanical computations (Cohen et al. (2012)). Alternatively, empirical potential functions (Torrens (2012)) can approximately capture this interaction while maintaining the minima associated with these structures. These potentials are developed relying only on two-, three- or four-body interactions and ignoring higher-order terms for computational efficiency. In this work, we rely on well-validated empirical potential to compute the energy of the different atomic structures. The major challenge in visualisation is the rough landscape featuring an enormous number of stable structures (local minima) and a large number of degrees of freedom associated with an atomic structure ($3N$ for an N-atom structure in 3 dimensional space). Previous studies have attempted to map the energy landscape using disconnectivity graphs for small clusters(Carr et al. (2016); Wales & Doye (1997); De Souza & Wales (2016)). Dimensionality reduction using principal component analysis has been used in studying the loss landscape of neural networks which also exhibits non-convexity(Gallagher & Downs (2003); Li et al. (2018)) While characterizing the energy landscape of actual material is challenging, several studies have focused on simple model systems. One of the classical systems extensively characterized includes the Lennard-Jones (LJ) system, which can be used to model noble gases (Tsai & Jordan (1993); Wales & Doye (1997); Malek & Mousseau (2000); Doye et al. (1999)). The energy of a system of N-atoms interacting through the LJ potential is given by:

$$U = \epsilon \sum_{i=1}^{N-1} \sum_{j>i}^{N} \left[\left(\frac{\sigma}{|x_{ij}|} \right)^{12} - \left(\frac{\sigma}{|x_{ij}|} \right)^{6} \right] \tag{1}$$

where $|x_{ij}| = |\mathbf{x_i} - \mathbf{x_j}|$ is the distance is between the atoms i and j, and ϵ and σ are constants depending on the atom types. By extrapolating the studies on small LJ structures, the scaling of minima with the number of atoms N can be obtained as $e^{(k_1+k_2N)}$ or $e^{(k_1+k_2N+k_3N^2)}$, where k_1, k_2 and k_3 are constants obtained by fitting (Wales & Doye (1997)). Thus, it becomes incredibly challenging to visualise the energy landscape of a system with thousands of atoms. The problem of visualising energy landscape can be stated as follows:

Problem: (Visualising Energy Landscape) *Let the energy function be $U(\theta) = U(\mathbf{x_1}, \mathbf{x_2}, ...\mathbf{x_N})$ for any configuration θ of an N-atom system, then our goal is to find δ and $\eta \in \mathbb{R}^{3N}$ such that $f(\alpha, \beta) = U(\theta^* + \alpha\delta + \beta\eta)$ approximates $U(\theta)$, for θ near θ^* using some interpolation parameters α and β .*

3 METHODOLGY

The goal is to find the directions δ and $\eta \in \mathbb{R}^{3N}$. As a first trial, one can pick two random unit directions sampled from $\mathbb{R}^{3N}$. However as we show later in section 4.1 it doesn't suffice our purpose. We propose to use principal component-based dimensionality reduction to pick directions that preserve the information about the actual landscape. First, we define the system and the energy function. In this work, we use a 100-atom binary Lennard Jones (LJ) system. Next, we sample some configurations from the energy landscape, either from Optimization trajectory or Molecular Dynamics simulation. From the sampled configurations, we pick one configuration as the origin (θ^*) and calculate the position vectors of all other configurations from the chosen origin θ^*. We then stack these vectors to form a matrix. Next, singular value decomposition is performed to find the principal components, along with the corresponding eigenvalues, λ_i's. Principal components corresponding to the two largest eigenvalues are chosen as projection directions δ and η. A grid of configurations around θ^* is formed using the interpolating parameters α and β. Finally, $f(\alpha, \beta)$ is evaluated to get the projected landscape. The complete stepwise methodology is presented in Figure 1

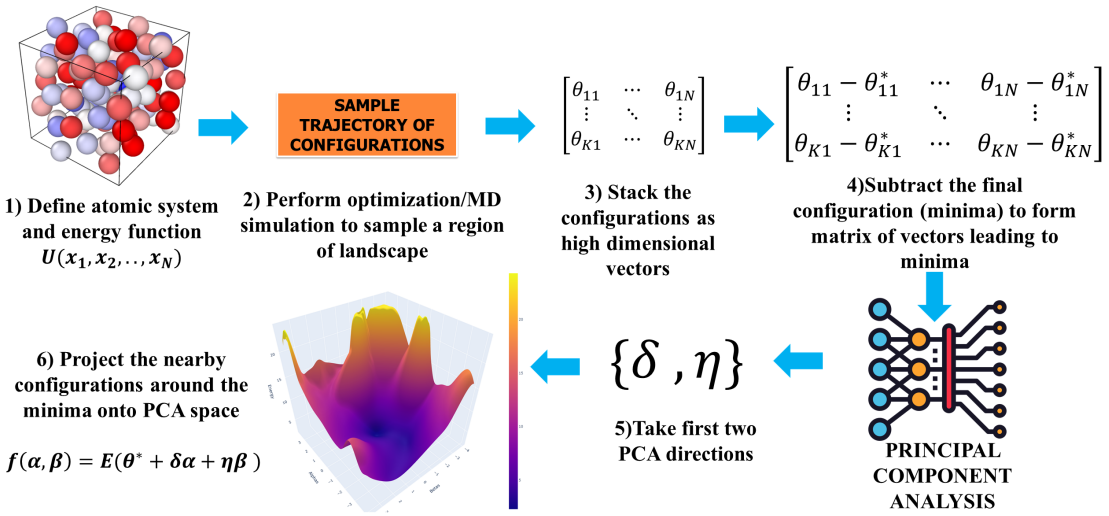


Figure 1: Steps followed in methodology for visualisation of energy landscapes

3.1 Validation of low dimensionality using explained Variance analysis

In order to evaluate whether the visualisation captures the actual landscape well enough, we calculate the explained variance defined as $\frac{\lambda_i}{\sum \lambda_i}$ where λ_i is the eigenvalue corresponding to the i'th principal component. It captures how well the i'th principal direction preserves the information of the sampled vectors from the actual landscape. In Figure 2 we show the cumulative explained variance as function of principal components for the configurations sampled from the gradient descent optimization trajectories. We observe that the first two principal components capture around 75% variance, indicating the visualization's usefulness.

3.2 Validation against actual curvatures

In this section, we ask ourselves whether the proposed visualization captures the curvatures in the actual landscape; a completely convex region has positive semi-definite Hessian, while non-convex regions have negative curvatures. Since the principal curvatures in the projected landscape are weighted averages of the curvatures of the actual landscape, any non-convexity present in the visualized landscape must also exist in the actual landscape; however, vice-versa may not be true. Thus, to verify whether convex regions of the visualized landscape correspond to the convex regions of the actual landscape, we study the curvature ratio obtained from the eigenvalues of the Hessian of the actual landscape. Curvature ratio is defined as $|\frac{\lambda_{min}}{\lambda_{max}}|$. This ratio is lower near convex regions as the near zero negative curvature eigenvalues become insignificant compared to the positive curvature eigenvalues. In Figure 3 we show that the convex looking regions in the visualised landscape do indeed correspond to the convex regions of the actual landscape. .

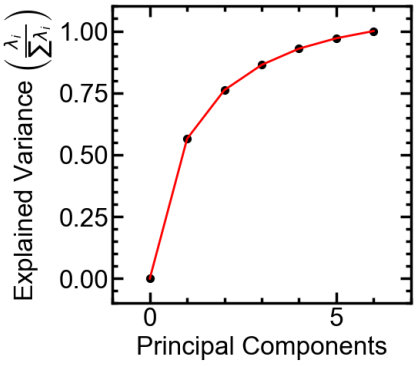


Figure 2: Explained Variance vs Principal Components for 100 atom LJ System, mean over 5 minimization trajectories. With only two component $\approx 75\%$ variance is captured

4 Experiments

4.1 Random Directions vs PCA Directions

In this section, we study the effect of choosing random directions as the projection directions δ and η. Figure 4 (a) shows the visualization of gradient descent optimization trajectory using random directions, and Figure 4(b) shows the visualization of the same trajectory using principal component

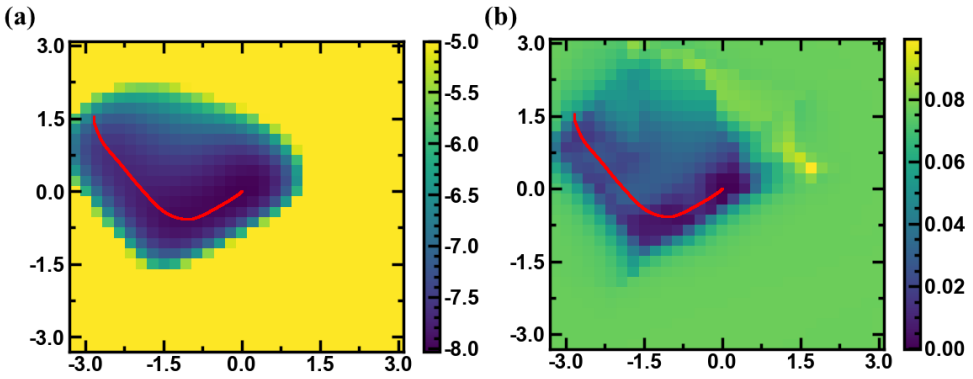
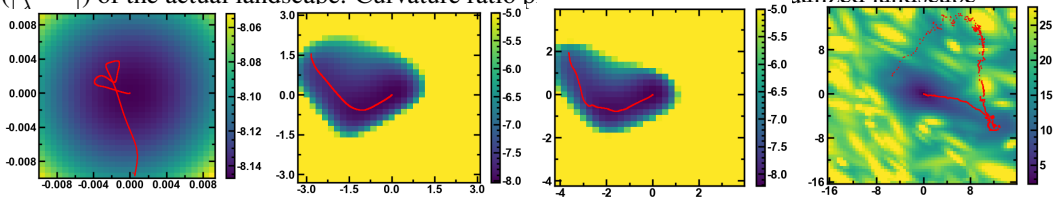


Figure 3: (a)2D Visualisation of Gradient descent optimization Trajectory, (b)Curvature Ratio ($|\frac{\lambda_{min}}{\lambda}|$) of the actual landscape. Curvature ratio plot matches with the visualized landscape



(a) Gradient Descent (Random Dir.)

(b) Gradient Descent (PCA Dir.)

(c) FIRE (PCA Dir.)

(d) VeLO (PCA Dir.)

Figure 4: Visualisation of (a) Gradient Descent with Random projection directions, (b) Gradient Descent with PCA based directions, (c) FIRE(Bitzek et al. (2006)) with PCA based directions, (d) Versatile Learned Optimizer (Metz et al. (2022)) with PCA based directions

analysis as proposed in section 3. We observe that random direction lead to chaotic trajectory in the visualised landscape, whereas a smooth trajectory is observed in case of PCA based directions. It shows that the PCA directions are indeed meaningful in representing optimization trajectories. Visualisations showing configurations sampled from MD Simulations are presented in App. Section A.1.

4.2 Machine learned vs Gradient Based Optimizer

In recent years, several machine-learned optimizers for atomic systems have been proposed (Merchant et al. (2021); Metz et al. (2022); Bihani et al. (2023)) exhibiting superior performance than traditional classical optimizers. However, the exact behavior of the machine-learned optimizer is not well understood. We apply our visualization methodology to see how the trajectory obtained using a machine-learned optimizer is different than the one obtained from the gradient-based optimizer. In Figure 4 (b), (c), and (d), we observe that trajectories obtained by VeLO (Metz et al. (2022)) follow a very different path than the gradient-based optimizers.It shows that machine-learned optimizer explores more in the energy landscape to find better minima.

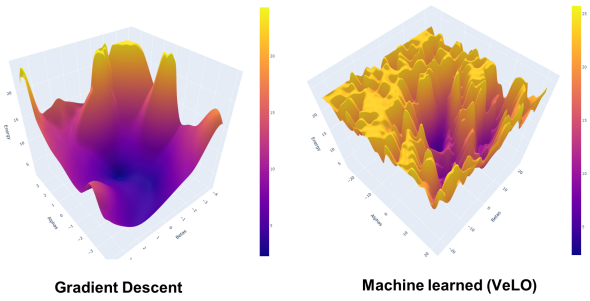


Figure 5: 3D Plot of the visualised landscape from Gradient Descent and Machine Learned trajectories

5 CONCLUSION

In this study, we addressed the challenge of visualizing the complex energy landscape of materials and proposed a method for low-dimensional projection of the energy landscape and validated our approach through Hessian analysis, demonstrating that the low-dimensional projection aligns well with the curvatures of the actual landscape. Additionally, we showcased the utility of our visualization method in providing insights into the behavior of various gradient-based and machine-learned optimizers. We believe that this visualization methodology will offer a fundamental comprehension of the energy landscape and accelerate the discovery and development of materials with targeted properties.

REFERENCES

Vaibhav Bihani, Sahil Manchanda, Srikanth Sastry, Sayan Ranu, and NM Anoop Krishnan. Stridernet: A graph reinforcement learning approach to optimize atomic structures on rough energy landscapes. In *International Conference on Machine Learning*, pp. 2431–2451. PMLR, 2023.

Erik Bitzek, Pekka Koskinen, Franz Gähler, Michael Moseler, and Peter Gumbsch. Structural relaxation made simple. *Physical review letters*, 97(17):170201, 2006.

Joanne M Carr, Dorian Mazauric, Frédéric Cazals, and David J Wales. Energy landscapes and persistent minima. *The Journal of chemical physics*, 144(5), 2016.

Aron J Cohen, Paula Mori-Sánchez, and Weitao Yang. Challenges for density functional theory. *Chemical reviews*, 112(1):289–320, 2012.

Vanessa K De Souza and David J Wales. The potential energy landscape for crystallisation of a lennard-jones fluid. *Journal of Statistical Mechanics: Theory and Experiment*, 2016(7):074001, 2016.

Jonathan PK Doye, Mark A Miller, and David J Wales. The double-funnel energy landscape of the 38-atom lennard-jones cluster. *The Journal of Chemical Physics*, 110(14):6896–6906, 1999.

Hans Frauenfelder, Stephen G Sligar, and Peter G Wolynes. The energy landscapes and motions of proteins. *Science*, 254(5038):1598–1603, 1991.

Marcus Gallagher and Tom Downs. Visualization of learning in multilayer perceptron networks using principal component analysis. *IEEE Transactions on Systems, Man, and Cybernetics, Part B (Cybernetics)*, 33(1):28–34, 2003.

Hao Li, Zheng Xu, Gavin Taylor, Christoph Studer, and Tom Goldstein. Visualizing the loss landscape of neural nets. *Advances in neural information processing systems*, 31, 2018.

Rachid Malek and Normand Mousseau. Dynamics of lennard-jones clusters: A characterization of the activation-relaxation technique. *Physical Review E*, 62(6):7723, 2000.

Sajid Mannan, Vaibhav Bihani, NM Anoop Krishnan, and John C Mauro. Navigating energy landscapes for materials discovery: Integrating modeling, simulation, and machine learning. *Materials Genome Engineering Advances*, pp. e25, 2024.

John C Mauro, Roger J Loucks, Arun K Varshneya, and Prabhat K Gupta. Enthalpy landscapes and the glass transition. *Scientific Modeling and Simulations*, pp. 241–281, 2009.

Amil Merchant, Luke Metz, Samuel S Schoenholz, and Ekin D Cubuk. Learn2hop: Learned optimization on rough landscapes. In *International Conference on Machine Learning*, pp. 7643–7653. PMLR, 2021.

Luke Metz, James Harrison, C Daniel Freeman, Amil Merchant, Lucas Beyer, James Bradbury, Naman Agrawal, Ben Poole, Igor Mordatch, Adam Roberts, et al. Velo: Training versatile learned optimizers by scaling up. *arXiv preprint arXiv:2211.09760*, 2022.

Iam Torrens. *Interatomic potentials*. Elsevier, 2012.

CJ Tsai and KD Jordan. Use of an eigenmode method to locate the stationary points on the potential energy surfaces of selected argon and water clusters. *The Journal of Physical Chemistry*, 97(43): 11227–11237, 1993.

David J Wales. Exploring energy landscapes. *Annual review of physical chemistry*, 69:401–425, 2018.

David J Wales and Jonathan PK Doye. Global optimization by basin-hopping and the lowest energy structures of lennard-jones clusters containing up to 110 atoms. *The Journal of Physical Chemistry A*, 101(28):5111–5116, 1997.

DJ Wales. *Energy Landscapes With Applications To Clusters, Biomolecules And Glasse*. Cambridge University Press, 2003.

A APPENDIX

A.1 VISUALISATIONS USING CONFIGURATIONS SAMPLED FROM MD SIMULATIONS

In this experiment we sample configurations from the MD simulation trajectories. First we sample a minimized configuration and excite the system by giving velocities to the atoms corresponding to low temperatures and allow the system to evolve in a constant energy(NVE) ensemble for large no. of steps. Next at every 100 steps we sample a configuration, minimize it using gradient descent and store the minimized configurations. We sample 10000 such configurations and use them to get the projection directions as explained in Section 3. Finally we obtain the projected landscape as shown in Figure 6

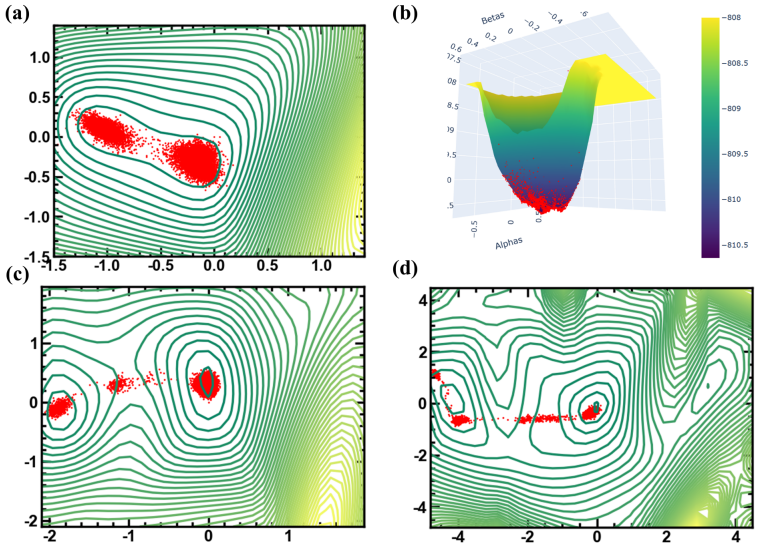


Figure 6: Visualisation of configurations sampled from MD simulation trajectories

We observe that the visualised landscape is able to capture clusters of different configurations present in different basins of the landscape. Further we observe that some configurations are trapped in the saddle points between two basins (Figure 6(c)). It shows that the proposed approach can be effectively used to study the charecteristics of energy landscape like basins, saddle points and transitions between them .