SUNCAT
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Accelerating *ab initio* simulations using surrogate machine learning models

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Summary:

Typically, first-principle calculations are computationally expensive. This problem is usually aggravated when modelling materials due to the large number of atoms involved in the models. Therefore, the design of robust and fast optimizers has always been a hot topic in this field. The desired algorithms must minimize the number of *ab initio* function calls as much as possible without compromising the accuracy of the simulated properties. Our approach is based on a machine learning surrogate model which allows to substantially reduce the number of function calls in the search of the optimal solution. Here, we present the evidence of the aforementioned acceleration for two of the most commonly encountered optimizations problems in materials science: energy minimization and transition-state search, such as structure optimizations and Nudged Elastic Band (NEB) calculations.

1. Machine Learning model:

- **CatLearn:** A package for building and testing **atomistic machine learning models**.
- Gaussian Progress Regression (**GPR**) routines are implemented in CatLearn.
- Predicted values and uncertainties of the **predictions** are obtained using GPR.
- Squared exponential **kernel** :

$$k_{SE}(x,x') = \sigma^2 \exp\left(-\frac{(x-x')^2}{2\ell^2}\right) \quad ; \quad \begin{array}{l} x/x' : \text{fingerprint} \\ \sigma : \text{scaling} \\ \ell : \text{length scale} \end{array}$$

- **CatLearn supports multiple covarying GP**. This model can learn from the observations and first derivative of the same observations (e.g. energies and forces). By including more information, the integral variance of the model is decreased systematically (**Fig. 1**). A detailed mathematical derivation can be found in Ref. 1.

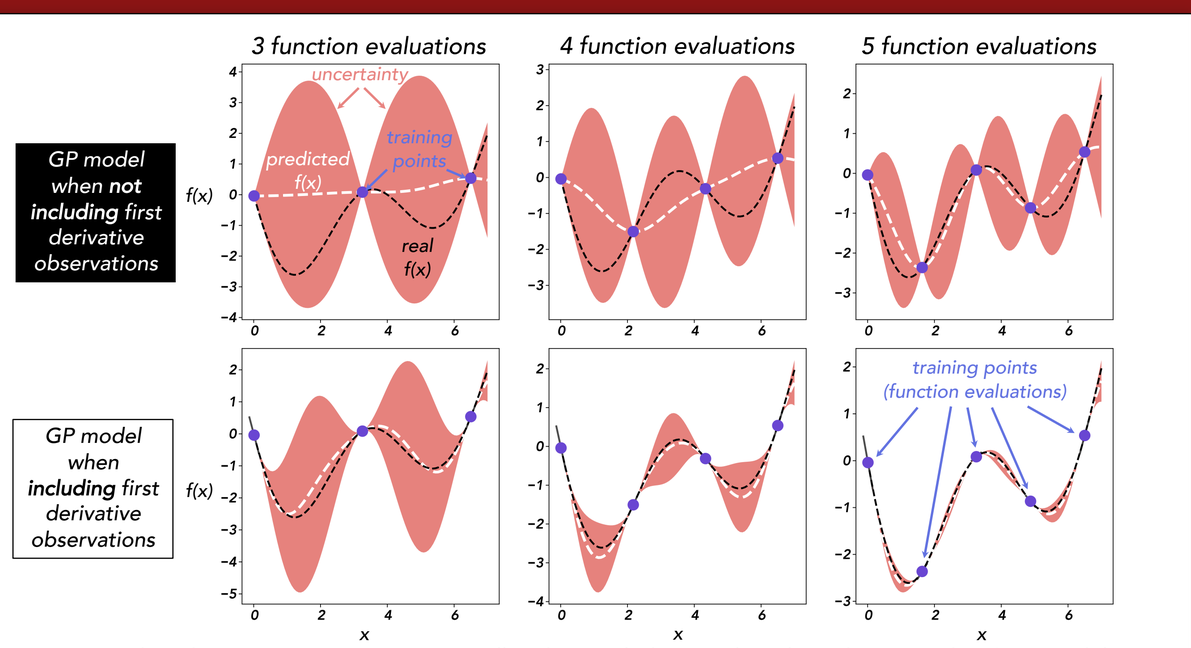

Fig. 1. Predicted mean improves systematically when including analytical gradients to the GPR model (compare first and second row).

2 Accelerating transition-state search:

Prototype model: Müller-Brown potential

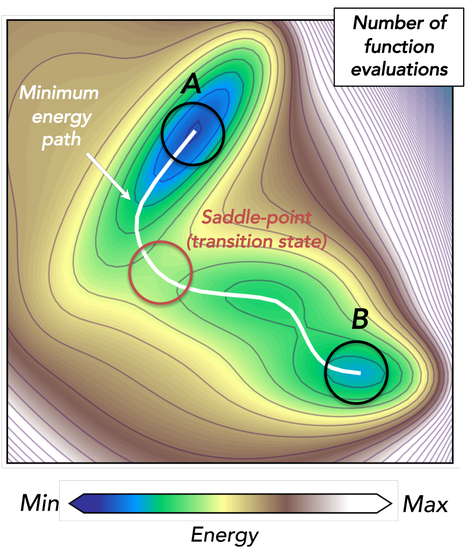

Fig. 2. Potential energy landscape for the Müller-Brown potential.

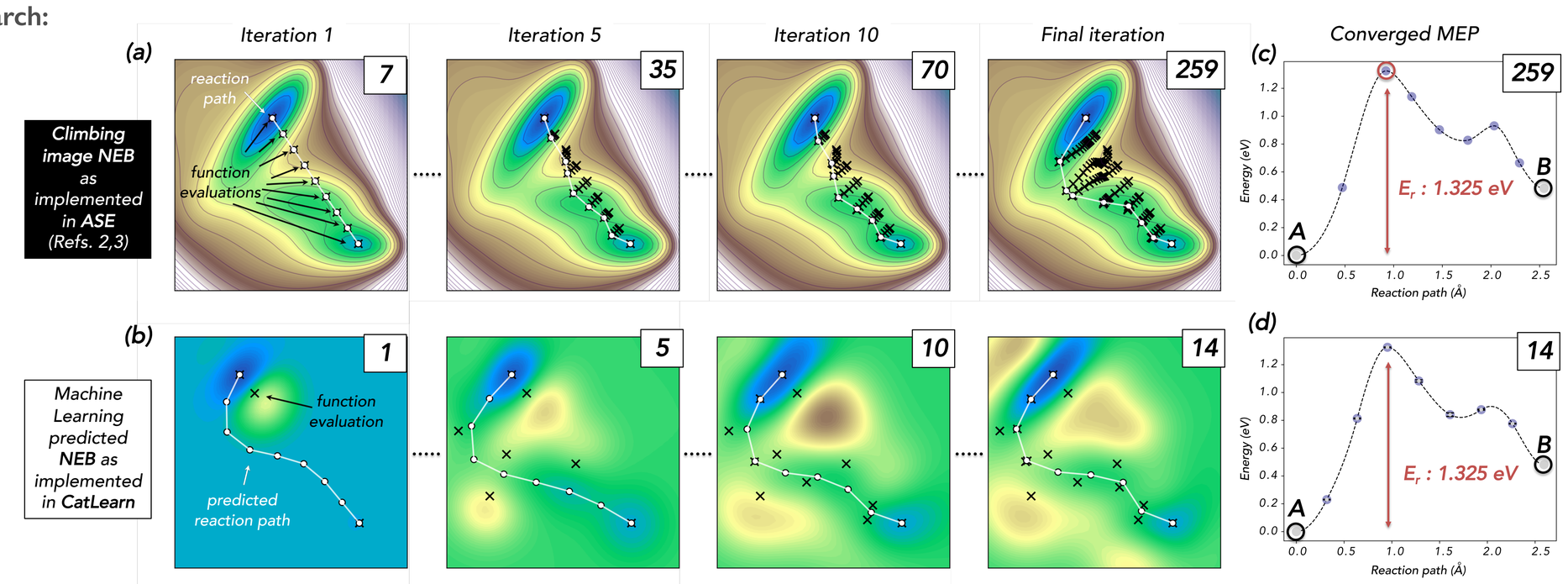

Fig. 3. (a) Evolution of the minimum energy path (MEP) for the Müller-Brown potential from the site A to B using Climbing Image NEB as implemented in ASE. (b) Evolution of the predicted potential energy surface and predicted NEB path obtained with our machine learning algorithm. (c, d) Converged minimum energy paths.

Objective:
Finding the minimum energy path (in **Fig. 2)** connecting the points A to B with the minimum number of function evaluations (energy and forces) possible.

Machine Learning algorithm:
We use a GPR model to build an "artificial" potential that can be used for obtaining a predicted NEB path. From this predicted path we evaluate the image with maximum uncertainty (only one image at the time) and we add it to the training list in order to improve the predicted potential energy surface (PES). After a few iterations the uncertainty of each image composing the NEB path goes bellow our stopping criteria (e.g. <5 meV), then we evaluate the top image to ensure that we found a saddle-point.

3. Accelerating and improving the robustness of structure optimization:

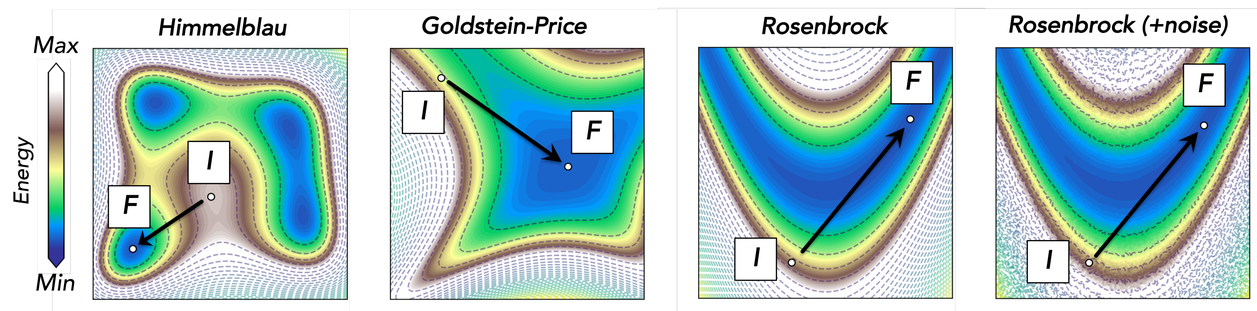

Fig. 4. Prototype models used to test the performance of the different algorithms (see **Table I**).
Note: in the Rosenbrock(+noise) function we added an artificial normally distributed random noise to the function.

Table I. Number of functions evaluations (energy and forces) performed by the Broyden-Fletcher-Goldfarb-Shanno (BFGS), Conjugate Gradients (CG), Nelder-Mead (as implemented in Scipy, Ref. 4) and CatLearn algorithms in order to achieve convergence ($\max(|f_i|) < 0.01$ a.u.) for the different prototype models shown in **Fig. 4**.

	Himmelblau	*Goldstein-Price*	*Rosenbrock*	*Rosenbrock (+noise)*
BFGS (L-BFGS-B)	19	17	29	Failed
CG	22	19	32	Failed
Nelder-Mead	82	56	183	Failed
CatLearn	**15**	**8**	**16**	**39**

Objective:
Finding the minimum of the prototype functions (in **Fig. 3**) from a common initial geometry (I) to the nearest local minima (F) employing the minimum number of function calls (energy and forces) possible.

Machine Learning algorithm:
We use a GPR model to build an "artificial" potential that we can minimize and predict an optimal value. We evaluate the image predicted from the surrogate model and we train the process including the evaluated image (expensive calculation), improving the predicted PES. After a few iterations the "next suggested training point" satisfies the convergence criteria.

4. Observations:

1) *Transition-state search.* Due to the fact that all images must be evaluated in each iteration, we observe a notoriously high number of function calls performed by the standard CI-NEB algorithm (259 energy and force evaluations, **Fig. 3a**). Our machine learning surrogate model allows to substantially reduce the number of functions calls in order to find the same saddle-point for the transition from A to B (only 14 energy and force evaluations, **Fig. 3b**).

2) *Structural optimization. Our GPR-based algorithm performs as well as the most popular quasi-Newton type algorithms in terms of number of function evaluations (**Table I**). In addition, the posterior mean obtained by the GPR allows to gain robustness when the function and its derivatives become noisy (**Fig. 4**). When dealing with noisy data the BFGS, CG and Nelder-Mead algorithms are incapable of reaching the desired convergence.*

* We have tested our algorithms with atomistic systems, including structures with a large number of atoms (>100 atoms per unit cell). The performance of the method holds for the prototype and the atomistic systems without sacrificing the accuracy of the results.

MADE IN SILICON VALLEY!!!!!!!
So, we have a code ready for action:

Please ask us for a live demonstration of any of the algorithms presented in this poster. In addition, we have a code available of the atomistic implementation of these algorithms working in a user-friendly fashion (similarly than the ones included in ASE).
Please visit us: https://github.com/SUNCAT-Center/CatLearn

References:

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