On-the-Fly Active Learning of Interpretable Bayesian Force Fields for Atomistic Rare Events: Supplementary Information

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I. DFT SETTINGS

The DFT settings used in this work are reported in Supplementary Table I below.

Material	Pseudopotential	K-points	Energy (Ry)	Density (Ry)
Al (Fig. 2)	Al.pbe-n-kjpaw-psl.1.0.0.UPF	7x7x7	29	143
Al (Figs. 3, 4a-c)	Al.pbe-n-kjpaw-psl.1.0.0.UPF	2x2x2	29	143
Al (Fig. 4d-f)	Al.pbe-n-kjpaw-psl.1.0.0.UPF	4x4x2	29	143
AgI	Ag.pbe-n-kjpaw_psl.1.0.0.UPF I.pbe-n-kjpaw_psl.1.0.0.UPF	Γ	45	181
C	C.pbe-n-kjpaw_psl.1.0.0.UPF	2x2x2	40	326
Si	Si.pbe-n-kjpaw_psl.1.0.0.UPF	Γ	44	175
Al_2O_3	Al.pbe-n-kjpaw-psl.1.0.0.UPF O.pbe-n-kjpaw_psl.1.0.0.UPF	Γ	47	323
NiTi	Ni.pz-n-rrkjus_psl.0.1.UPF Ti.pz-spn-rrkjus_psl.1.0.0.UPF	2x2x2	52	576
BN	B.pbe-n-kjpaw_psl.1.0.0.UPF N.pbe-n-kjpaw_psl.1.0.0.UPF	Γ	44	325

TABLE I. Details of the DFT calculations performed in this work. All pseudopotentials are available in the Quantum Espresso pseudopotential library [1, 2].

II. KERNEL EVALUATION COST

To provide an estimate of the computational cost of evaluating forces with our Python-based GP models before mapping them to cubic splines, we provide empirical wall times of 2- and 2+3-body kernel evaluations and GP force and variance predictions in Supplementary Table II below. The test was performed with the 2- and 2+3-body aluminum melt GP models presented in Fig. 3 of the main text. All tests were performed on the same atomic environment ρ_{test} taken from the training set of the GP, which contained 42 atoms inside the 2-body cutoff sphere and 18 atoms inside the 3-body cutoff sphere. We report in the first two rows the wall time of evaluating $k_{y,z}(\rho_{\text{test}}, \rho_{\text{test}})$ for the 2- and 2+3-body kernels on a single CPU core, showing that the 2+3-body kernel requires about an order of magnitude more wall time due to the double loop over neighbors that is required. The next two rows report the cost of predicting the x-component of the force on ρ_{test} along with the epistemic uncertainty σ_{ix} on this force component. The time is approximately a factor of $93 \times 3 = 279$ larger than the corresponding kernel evaluation times, due to the loop over training labels required in the evaluation of the GP mean and variance (Eq. (5) of the main text). We also report in the final two rows the cost of evaluating the mean prediction alone, which is only slightly less than the cost of evaluating both the mean and variance. This is because the majority of the computation time in both cases is spent evaluating the kernel vector $\bar{k}_{i\alpha}$ in Eq. (5).

Computed Quantity	Time (ms)
2-body kernel	0.039
2+3-body kernel	0.46
2-body prediction (M+V)	11.36
2+3-body prediction (M+V)	138.72
2-body prediction (M)	10.90
2+3-body prediction (M)	136.71

TABLE II. Wall times of kernel evaluations and GP force and variance predictions. Reported wall times were averaged over 1000 evaluations.

III. CUBIC SPLINE MODELS

In Supplementary Table III, we report the grid of control points used to construct the spline-based tabulated force fields discussed in the main text.

Model	2-body	3-body
High entropy alloy (Fig. 2f)	100	$30 \times 30 \times 30$
2-body Aluminum (Fig. 3)	100	-
$2 + 3$-body Aluminum (Fig. 3)	100	$15 \times 15 \times 15$
Silver iodide (Fig. 5)	64	$15 \times 15 \times 15$

TABLE III. Cubic spline grids of the mapped force fields discussed in the main text.

IV. DERIVATIVE KERNEL EXPRESSIONS

Expressions used to compute the three-body force kernel defined in Eq. (3) of the main text are reported in Supplementary Table IV.

	Quantity	Definition
Energy Kernel	$k_3(\mathbf{d}_{i,i_1,i_2}, \mathbf{d}_{j,j_1,j_2})$	$\sigma_{s,3}^2 k_{\mathrm{SE}}(\mathbf{d}_{i,i_1,i_2}, \mathbf{d}_{j,j_1,j_2}) f_{\mathrm{cut}}(\mathbf{d}_{i,i_1,i_2}) f_{\mathrm{cut}}(\mathbf{d}_{j,j_1,j_2})$
	$k_{\mathrm{SE}}(\mathbf{d}_{i,i_1,i_2}, \mathbf{d}_{j,j_1,j_2})$	$\exp\left(-\frac{\lVert\mathbf{d}_{i,i_1,i_2}-\mathbf{d}_{j,j_1,j_2}\rVert^2}{2\ell^2}\right)$
	$\mathbf{d}_{j,j_1,j_2}$	$(r_{i,i_1}, r_{i,i_2}, r_{i_1,i_2})$
	$f_{\mathrm{cut}}(\mathbf{d}_{i,i_1,i_2})$	$f(r_{i,i_1})f(r_{i,i_2})f(r_{i_1,i_2})$
	$f(r)$	$(r - r_{\mathrm{cut}})^2$
Force Kernel	$\frac{\partial^2 k_3}{\partial \vec{r}_{i\alpha} \partial \vec{r}_{j\beta}}$	$\sigma_{s,3}^2(k_0 + k_1 + k_2 + k_3)$
	k_0	$k_{\mathrm{SE}} \frac{\partial f_{\mathrm{cut}}(\mathbf{d}_{i,i_1,i_2})}{\partial \vec{r}_{i\alpha}} \frac{\partial f_{\mathrm{cut}}(\mathbf{d}_{j,j_1,j_2})}{\partial \vec{r}_{j\beta}}$
	k_1	$\frac{\partial k_{\mathrm{SE}}}{\partial \vec{r}_{i\alpha}} f_{\mathrm{cut}}(\mathbf{d}_{i,i_1,i_2}) \frac{\partial f_{\mathrm{cut}}(\mathbf{d}_{j,j_1,j_2})}{\partial \vec{r}_{j\beta}}$
	k_2	$\frac{\partial k_{\mathrm{SE}}}{\partial \vec{r}_{j\beta}} \frac{\partial f_{\mathrm{cut}}(\mathbf{d}_{i,i_1,i_2})}{\partial \vec{r}_{i\alpha}} f_{\mathrm{cut}}(\mathbf{d}_{j,j_1,j_2})$
	k_3	$\frac{\partial^2 k_{\mathrm{SE}}}{\partial \vec{r}_{i\alpha} \partial \vec{r}_{j\beta}} f_{\mathrm{cut}}(\mathbf{d}_{i,i_1,i_2}) f_{\mathrm{cut}}(\mathbf{d}_{j,j_1,j_2})$
	$\frac{\partial k_{\mathrm{SE}}}{\partial \vec{r}_{i\alpha}}$	$\frac{k_{\mathrm{SE}} B_1}{\ell^2}$
	B_1	$\frac{(r_{i,i_1}-r_{j,j_1})(\vec{r}_{i_1\alpha}-\vec{r}_{i\alpha})}{r_{i,i_1}} + \frac{(r_{i,i_2}-r_{j,j_2})(\vec{r}_{i_2\alpha}-\vec{r}_{i\alpha})}{r_{i,i_2}}$
	$\frac{\partial k_{\mathrm{SE}}}{\partial \vec{r}_{j\beta}}$	$-\frac{k_{\mathrm{SE}} B_2}{\ell^2}$
	B_2	$\frac{(r_{i,i_1}-r_{j,j_1})(\vec{r}_{j_1\beta}-\vec{r}_{j\beta})}{r_{j,j_1}} + \frac{(r_{i,i_2}-r_{j,j_2})(\vec{r}_{j_2\beta}-\vec{r}_{j\beta})}{r_{j,j_2}}$
	$\frac{\partial^2 k_{\mathrm{SE}}}{\partial \vec{r}_{i\alpha} \vec{r}_{j\beta}}$	$\frac{k_{\mathrm{SE}}}{\ell^4}\left(A\ell^2 - B_1 B_2\right)$
	A	$\frac{(\vec{r}_{i_1\alpha}-\vec{r}_{i\alpha})(\vec{r}_{j_1\beta}-\vec{r}_{j\beta})}{r_{i,i_1} r_{j,j_1}} + \frac{(\vec{r}_{i_2\alpha}-\vec{r}_{i\alpha})(\vec{r}_{j_2\beta}-\vec{r}_{j\beta})}{r_{i,i_2} r_{j,j_2}}$
ℓ Derivative	$\frac{\partial^3 k_3}{\partial \ell \partial \vec{r}_{i\alpha} \partial \vec{r}_{j\beta}}$	$\sigma_{s,3}^2\left(\frac{\partial k_0}{\partial \ell} + \frac{\partial k_1}{\partial \ell} + \frac{\partial k_2}{\partial \ell} + \frac{\partial k_3}{\partial \ell}\right)$
	$\frac{\partial k_{\mathrm{SE}}}{\partial \ell}$	$\frac{k_{\mathrm{SE}} \lVert\mathbf{d}_{i,i_1,i_2}-\mathbf{d}_{j,j_1,j_2}\rVert^2}{l^3}$
	$\frac{\partial^2 k_{\mathrm{SE}}}{\partial \ell \partial \vec{r}_{i\alpha}}$	$B_1\left(\frac{1}{\ell^2}\frac{\partial k_{\mathrm{SE}}}{\partial \ell} - \frac{2k_{\mathrm{SE}}}{\ell^3}\right)$
	$\frac{\partial^2 k_{\mathrm{SE}}}{\partial \ell \partial \vec{r}_{j\beta}}$	$-B_2\left(\frac{1}{\ell^2}\frac{\partial k_{\mathrm{SE}}}{\partial \ell} - \frac{2k_{\mathrm{SE}}}{\ell^3}\right)$
	$\frac{\partial^3 k_{\mathrm{SE}}}{\partial \ell \partial \vec{r}_{i\alpha} \partial \vec{r}_{j\beta}}$	$\left(A\ell^2 - B_1 B_2\right)\left(\frac{\partial k_{\mathrm{SE}}}{\partial \ell}\frac{1}{\ell^4} - \frac{4k_{\mathrm{SE}}}{\ell^5}\right) + \frac{2k_{\mathrm{SE}} A}{\ell^3}$
σ Derivative	$\frac{\partial^3 k_3}{\partial \sigma_{s,3} \partial \vec{r}_{i\alpha} \partial \vec{r}_{j\beta}}$	$2\sigma_{s,3}(k_0 + k_1 + k_2 + k_3)$

TABLE IV. Quantities used to calculate the three-body force kernel and its derivatives with respect to the hyperparameters ℓ and σ, which are required in the calculation of the log marginal likelihood.

V. REFERENCES

[1] Andrea Dal Corso, "Pseudopotentials periodic table: From h to pu," Computational Materials Science **95**, 337–350 (2014).

[2] `https://www.quantum-espresso.org/pseudopotentials`.