I. TAYLOR EXPANSION POTENTIAL

Let C denotes a 3×3 matrix composed of the lattice vector (column vector) $\boldsymbol{a}_1$, $\boldsymbol{a}_2$, and $\boldsymbol{a}_3$ defined as

$$C = (\boldsymbol{a}_1, \boldsymbol{a}_2, \boldsymbol{a}_3). \tag{1}$$

The Taylor expansion potential (TEP) is defined as

$$U_{\rm TEP} = \sum_{n=0}^{\infty} U_n = U_0 + U_1 + U_2 + U_3 + \cdots, \tag{2}$$

where

$$U_n = \frac{1}{n!} \sum_{\{\ell,\kappa,\mu\}} \Phi_{\mu_1 \ldots \mu_n}(\ell_1\kappa_1; \ldots; \ell_n\kappa_n)\, u_{\mu_1}(\ell_1\kappa_1) \cdots u_{\mu_n}(\ell_n\kappa_n). \tag{3}$$

Here, the atomic displacements are defined in the Cartesian coordinate. Let us denote a column vector comprising the N irreducible interatomic force constants (IFCs) as $\boldsymbol{\Phi}$. Then, the TEP potential is symbolically written as

$$U_{\rm TEP} = \boldsymbol{b}^T \boldsymbol{\Phi}. \tag{4}$$

The atomic forces in the Cartesian coordinate is given as

$$\boldsymbol{F}_{\rm TEP}(\ell\kappa) = -\frac{\partial U_{\rm TEP}}{\partial \boldsymbol{R}(\ell\kappa)} = -\frac{\partial \boldsymbol{b}^T}{\partial \boldsymbol{u}(\ell\kappa)} \boldsymbol{\Phi} = A_{\ell\kappa} \boldsymbol{\Phi}. \tag{5}$$

Now, let us consider the IFCs in the fractional coordinate $\tilde{\boldsymbol{R}}(\ell\kappa)$ instead of the Cartesian coordinate $\boldsymbol{R}(\ell\kappa)$. Since the Cartesian components and the fractional components are related with each other as

$$\boldsymbol{R}(\ell\kappa) = C\tilde{\boldsymbol{R}}(\ell\kappa), \tag{6}$$

it is easy to show the following conversion holds:

$$\tilde{\Phi}_{\mu_1' \ldots \mu_n'}(\ell_1\kappa_1; \ldots; \ell_n\kappa_n) = \sum_{\{\mu\}} C_{\mu_1\mu_1'} \cdots C_{\mu_n\mu_n'} \Phi_{\mu_1 \ldots \mu_n}(\ell_1\kappa_1; \ldots; \ell_n\kappa_n), \tag{7}$$

where $\tilde{\Phi}$ is the IFCs defined in the fractional coordinate. The above expression can be inverted as

$$\Phi_{\mu_1 \ldots \mu_n}(\ell_1\kappa_1; \ldots; \ell_n\kappa_n) = \sum_{\{\mu'\}} (C^{-1})_{\mu_1'\mu_1} \cdots (C^{-1})_{\mu_n'\mu_n} \tilde{\Phi}_{\mu_1' \ldots \mu_n'}(\ell_1\kappa_1; \ldots; \ell_n\kappa_n). \tag{8}$$

By introducing a column vector $\tilde{\mathbf{\Phi}}$, the TEP energy and forces are written as follows:

$$U_{\mathrm{TEP}} = \tilde{\boldsymbol{b}}^T \tilde{\mathbf{\Phi}}, \tag{9}$$

$$\tilde{\boldsymbol{F}}_{\mathrm{TEP}}(\ell\kappa) = -\frac{\partial U_{\mathrm{TEP}}}{\partial \tilde{\boldsymbol{R}}(\ell\kappa)} = -\frac{\partial \tilde{\boldsymbol{b}}^T}{\partial \tilde{\boldsymbol{u}}(\ell\kappa)} \tilde{\mathbf{\Phi}} = \tilde{A}_{\ell\kappa} \tilde{\mathbf{\Phi}} = C \boldsymbol{F}_{\mathrm{TEP}}(\ell\kappa). \tag{10}$$

So far, the matrices A and $\tilde{A}$ ($\in \mathbb{R}^{3\times N}$) are defined for each atom $\ell\kappa$. Now, we concatenate the columb vectors $\{\boldsymbol{F}(\ell\kappa)\}$ and matrices $\{A_{\ell\kappa}\}$ for all atoms in the supercell as follows:

$$\boldsymbol{F}^T = [\boldsymbol{F}_{11}^T, \boldsymbol{F}_{12}^T, \ldots, \boldsymbol{F}_{N_\ell N_\kappa}^T], \tag{11}$$

$$A^T = [A_{11}^T, A_{12}^T, \ldots, A_{N_\ell N_\kappa}^T]. \tag{12}$$

Here, N_ℓ is the number of unit cells in a supercell and N_κ is the number of atoms in each unit cell. For the brevity of the notation, we simply use $N_s = N_\ell N_\kappa$. By using the vector $\boldsymbol{F} \in \mathbb{R}^{3N_s\times 1}$ and matrix $A \in \mathbb{R}^{3N_s\times N}$, we obtain

$$\boldsymbol{F} = A\mathbf{\Phi}, \tag{13}$$

$$\tilde{\boldsymbol{F}} = \tilde{A}\tilde{\mathbf{\Phi}}. \tag{14}$$

Atomic forces in the Cartesian coordinate and the fractional coordinates are related to each other via the matrix C as in Eq. (10). Therefore, the columb vectors $\boldsymbol{F}$ and $\tilde{\boldsymbol{F}}$ are related as follows:

$$\tilde{\boldsymbol{F}} = \mathcal{C}\boldsymbol{F}. \tag{15}$$

Here, $\mathcal{C} \in \mathbb{R}^{3N_s\times 3N_s}$ is the block diagonal matrix defined as

$$\mathcal{C} = \begin{bmatrix} C & 0 & \cdots & 0 \\ 0 & C & \cdots & 0 \\ \vdots & \vdots & \ddots & 0 \\ 0 & 0 & \cdots & C \end{bmatrix}. \tag{16}$$

II. LINEAR REGRESSION

A. Ordinary least squares

In the ordinary least squares (OLS), we estimate the force constant vector by solving the following optimization problem:

$$\mathbf{\Phi}_{\mathrm{OLS}} = \arg\min_{\mathbf{\Phi}} \frac{1}{2N_d} \|\boldsymbol{\mathcal{F}}_{\mathrm{DFT}} - \mathbb{A}\mathbf{\Phi}\|_2^2. \tag{17}$$

Here, $\boldsymbol{\mathcal{F}}^T = [\boldsymbol{F}^T(\boldsymbol{u}_1), \boldsymbol{F}^T(\boldsymbol{u}_2), \ldots, \boldsymbol{F}^T(\boldsymbol{u}_{N_d})]$ and $\mathbb{A}^T = [A^T(\boldsymbol{u}_1), A^T(\boldsymbol{u}_2), \ldots, A^T(\boldsymbol{u}_{N_d})]$, where N_d is the number of displacement patterns. In other words, the force constants are determined in such a way that the DFT atomic forces in the Cartesian coordinate are best represented by the TEP. The above problem can be solved easily via the singular value decomposition or QR decomposition.

Now, let us derive the OLS optimization problem for $\tilde{\boldsymbol{\Phi}}$, which is the force constant vector in the fractional coordinate. Since we need to minimize the residual of atomic forces in the Cartesian coordinate instead of the fractional coordinate, we can simply use Eq. (17). First, we can easily show

$$A\boldsymbol{\Phi} = \mathcal{C}^{-1}\tilde{A}\tilde{\boldsymbol{\Phi}} = B\tilde{\boldsymbol{\Phi}}. \tag{18}$$

Therefore, we can convert Eq. (17) as

$$\tilde{\boldsymbol{\Phi}}_{\mathrm{OLS}} = \arg\min_{\tilde{\boldsymbol{\Phi}}} \frac{1}{2N_d}\|\boldsymbol{\mathcal{F}}_{\mathrm{DFT}} - \mathbb{B}\tilde{\boldsymbol{\Phi}}\|_2^2, \tag{19}$$

where $\mathbb{B}^T = [B^T(\boldsymbol{u}_1), B^T(\boldsymbol{u}_2), \ldots, B^T(\boldsymbol{u}_{N_d})]$. We note that the OLS solutions $\boldsymbol{\Phi}_{\mathrm{OLS}}$ and $\tilde{\boldsymbol{\Phi}}_{\mathrm{OLS}}$ are related with each other via Eq. (8). Therefore, the OLS solution does not depend on the basis of force constants.

B. Elastic net

In the elastic-net (enet) optimization, we solve the following problem:

$$\boldsymbol{\Phi}_{\mathrm{enet}} = \arg\min_{\boldsymbol{\Phi}} \frac{1}{2N_d}\|\boldsymbol{\mathcal{F}}_{\mathrm{DFT}} - \mathbb{A}\boldsymbol{\Phi}\|_2^2 + \alpha\beta\|\boldsymbol{\Phi}\|_1 + \frac{1}{2}\alpha(1-\beta)\|\boldsymbol{\Phi}\|_2^2. \tag{20}$$

From the similarity of the argument in the previous subsection, we may use the following problem for $\tilde{\boldsymbol{\Phi}}$:

$$\tilde{\boldsymbol{\Phi}}_{\mathrm{enet}} = \arg\min_{\tilde{\boldsymbol{\Phi}}} \frac{1}{2N_d}\|\boldsymbol{\mathcal{F}}_{\mathrm{DFT}} - \mathbb{B}\tilde{\boldsymbol{\Phi}}\|_2^2 + \alpha\beta\|\tilde{\boldsymbol{\Phi}}\|_1 + \frac{1}{2}\alpha(1-\beta)\|\tilde{\boldsymbol{\Phi}}\|_2^2. \tag{21}$$

We note that the enet solutions $\boldsymbol{\Phi}_{\mathrm{enet}}$ and $\tilde{\boldsymbol{\Phi}}_{\mathrm{enet}}$ cannot be transformed to each other via Eq. (8) due to the presence of the L_1 and L_2 regularization terms.

III. TRANSLATIONAL INVARIANCE