Supporting Information for

Coarse-graining of Imaginary Time Feynman Path Integrals: Inclusion of Intramolecular Interactions and Bottom-up Force-matching

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Functional Details of the Coupling Terms in Eq. (11)

The coupling matrices and scalar from Eq. (11) can be written as

$$\boldsymbol{K}_{Q\bar{q}} = \frac{\mathbb{L}^{-1}}{\beta} - \frac{\boldsymbol{M}}{2} - \sum_{Z} D^2 V_z(\bar{\boldsymbol{q}}) \tag{S1a}$$

$$\boldsymbol{K}_{\Delta q^2} = \frac{\boldsymbol{M}}{24} + \frac{\boldsymbol{\Sigma}}{\beta} - \frac{\beta}{4}\boldsymbol{C} + \frac{1}{12}\sum_{Z} D^2 V_z(\bar{\boldsymbol{q}}) \tag{S1b}$$

$$K_0 = \frac{1}{2\beta} ln\left[\left(\prod_{i=1}^{N} \frac{4m_i}{\beta\hbar^2}\right) det(\boldsymbol{\Omega} + \boldsymbol{\Omega_2} + \boldsymbol{I}) det(\boldsymbol{L})\right]. \tag{S1c}$$

For two-body terms

$$\sigma_{ij} = \sqrt{\frac{\hbar^2\beta}{\sqrt{m_i m_j}}} \tag{S2a}$$

$$\sigma_i = \sqrt{\frac{\hbar^2\beta}{m_i}} \tag{S2b}$$

$$\gamma_{ij} = \left(\frac{m_j}{m_i}\right)^{\frac{1}{4}}, \tag{S2c}$$

we define two N dimensional square block matrices whose matrix elements are infinite dimensional diagonal matrix $\boldsymbol{\Omega}_1$ and $\boldsymbol{\Omega}_2$. The indices i and j are for block elements and the indices k and l are for within the element matrices. Their elements are written as

$$(\boldsymbol{\Omega}_1)_{ij,kl} = \begin{cases} \delta_{kl} \sum_{m \neq i}^{N} \begin{pmatrix} \frac{\beta \sigma_{im}{}^2}{2(k\pi)^2} \overline{V''_{im}} \\ +(\gamma_{im}{}^2 - 1) \end{pmatrix} & (i = j) \\ -\delta_{kl} \frac{\beta \sigma_{ij}{}^2}{2(k\pi)^2} \overline{V''_{ij}} & (i \neq j) \end{cases} \tag{S3}$$

for

$$\overline{V''_{ij}} = V''_{ij}[\bar{q}_i - \bar{q}_j] + V''_{ij}[\bar{q}_j - \bar{q}_i] \tag{S4}$$

and

$$(\boldsymbol{\Omega}_2)_{ij,kl} = \sum_z \frac{2\beta}{\pi^2} \frac{\sigma_{ij}{}^2}{kl} \int_0^1 du \left(D^2 V_{z,ij} \big|_c \right) sin(k\pi u) sin(l\pi u) \tag{S5}$$

for $D^2 V_{z,ij}\big|_c$ which is the ith row jth column element of the Hessian matrix of V_z evaluated at the classical path (linear line between the beginning and the end of the path).

For given $\boldsymbol{\Omega}_1$ and $\boldsymbol{\Omega}_2$, and the identity matrix $\boldsymbol{I}$ of same dimensionality, we define a matrix $\boldsymbol{\Lambda}$ given by

$$\boldsymbol{\Lambda} = (\boldsymbol{\Omega}_1 + \boldsymbol{\Omega}_2 + \boldsymbol{I})^{-1} \tag{S6}$$

with ith row jth column block element $\boldsymbol{\Lambda}_{ij}$. Moreover, for infinite dimensional column vectors $\boldsymbol{b}_\xi$ and $\boldsymbol{b}_\lambda$ with elements

$$\left(\boldsymbol{b}_\xi\right)_k = -\frac{2\sqrt{2}}{\pi^2} \frac{odd(k)}{k^2} \tag{S7a}$$

$$(\boldsymbol{b}_\lambda)_k = \frac{\sqrt{2}}{\pi^2} \frac{1 - odd(k)}{k^2} \tag{S7b}$$

$$odd(k) = \begin{cases} 1 \ (k = odd) \\ 0 \ (k = even) \end{cases} \tag{S7c}$$

we can define two N dimensional square matrices $\boldsymbol{L}$ and $\boldsymbol{R}$. Their ith row and jth column elements are specified by

$$(\boldsymbol{L})_{ij} = \sigma_i \sigma_j \left(\boldsymbol{b}_\xi{}^T \boldsymbol{\Lambda}_{ij} \boldsymbol{b}_\xi \right) \tag{S8a}$$

$$(\boldsymbol{R})_{ij} = \sigma_i \sigma_j \left(\boldsymbol{b}_\lambda{}^T \boldsymbol{\Lambda}_{ij} \boldsymbol{b}_\lambda \right). \tag{S8b}$$

The ith row and jth column elements for the N dimensional square matrices $\boldsymbol{\Sigma}$ and $\boldsymbol{M}$ are written as

$$(\boldsymbol{\Sigma})_{ij} = \frac{\delta_{ij}}{{\sigma_i}^2} \tag{S9a}$$

$$(\boldsymbol{M})_{ij} = \begin{cases} \sum_{m \neq i}^{N} \overline{V''_{im}}\ (for\ i = j) \\ -2V''_{ij}(\bar{q}_i - \bar{q}_j)\ (for\ i \neq j) \end{cases}. \tag{S9b}$$

The N dimensional square matrix $\sum_Z D^2 V_z(\overline{\boldsymbol{q}})$ is a Hessian matrix of V_z evaluated at $\overline{\boldsymbol{q}}$. Lastly, the N dimensional square matrix $\boldsymbol{C}$ satisfies the relation

$$\Delta\boldsymbol{q}^T \boldsymbol{C} \Delta\boldsymbol{q} = \left(\boldsymbol{G} + 2\mathcal{E}^{(2)}\right)^T \boldsymbol{R}\left(\boldsymbol{G} + 2\mathcal{E}^{(2)}\right) \tag{S10}$$

for N dimensional column vectors $\boldsymbol{G}$ and $\mathcal{E}^{(2)}$ with ith row element

$$(\boldsymbol{G})_i = \sum_{\substack{j=1 \\ j \neq i}}^{N} (\Delta q_i - \Delta q_j)\overline{V''_{ij}} \tag{S11a}$$

$$\left(\mathcal{E}^{(2)}\right)_i = \sum_z D^2 V_{z,i}\Big|_{\overline{\boldsymbol{q}}} \cdot \Delta\boldsymbol{q} \tag{S11b}$$

where $D^2 V_{z,i}\Big|_{\overline{\boldsymbol{q}}}$ is the ith row of the Hessian matrix of V_z evaluated with respect to $\overline{\boldsymbol{q}}$.

Derivation of Eq. (11)

We present a derivation of Eq. (11) of the main text in this section. This section is a generalized version of derivation for Eq. (12) of our previous work.[1] Throughout the current derivation, we will refer to the previous derivation.

An alternative representation of the imaginary path using Fourier decomposition can be written as

$$q_i(u) = q_i(0) + \left(q_i(1) - q_i(0)\right)u + \frac{\sqrt{2}}{\pi}\sigma_i \sum_{k=1}^{\infty} a_{i,k} sin(k\pi u)/k \tag{S12}$$

where $\sigma_i = \sqrt{\hbar^2 \beta / m_i}$ where m_i is the mass associated with the ith particle or ring polymer, $a_{i,k}$ are the path parameter where i indexes the ring polymer number and k indexes the Fourier decomposition.[2] The variable u ranges from 0 to 1. $q_i(0)$ is the beginning of the path and $q_i(1)$ is the end of the path. For notational simplicity, we decompose Eq. (S1) into two parts: the classical ($q_{C,i}(u)$) and the quantum path ($q_{Q,i}(u)$) such that

$$q_{C,i}(u) = q_i(0) + \big(q_i(1) - q_i(0)\big)u \tag{S13}$$

$$q_{Q,i}(u) = \frac{\sqrt{2}\sigma_i}{\pi}\sum_{n=1}^{\infty} a_{i,n}\sin(n\pi u)/n. \tag{S13}$$

Note that the classical path is a linear line between the beginning and the end of the imaginary path while the quantum path represents a delocalization from the said classical path as an infinite sum of sine functions. Using Eq. (S12), the exact expression of the many-body CG potential $V_{CG,N}(\boldsymbol{q},\boldsymbol{Q},\boldsymbol{q}')$ can be written as

$$e^{-\beta V_{CG,N}(\boldsymbol{q},\boldsymbol{Q},\boldsymbol{q}')} = \left(\prod_{i=1}^{N}\sigma_i/\sqrt{8\pi}\right) exp\left(-\frac{1}{2}\Delta\boldsymbol{q}^T\boldsymbol{\Sigma}\Delta\boldsymbol{q}\right)$$

$$\times\int\frac{d\xi d\boldsymbol{a}}{\sqrt{2\pi}}\begin{bmatrix} exp\left(-\frac{1}{2}\boldsymbol{a}^2\right) exp\left(-\beta\int_0^1 du\, V\big(q_1(u),\ldots,q_N(u)\big)\right) \\ exp\big(i\boldsymbol{\xi}\cdot(\boldsymbol{Q}-\overline{\boldsymbol{q}})\big)\, exp\left(-i\frac{2\sqrt{2}}{\pi^2}\sum_{m=1}^{N}\sum_{k=1,3,5,\ldots}^{\infty}\sigma_m\xi_m a_{m,k}/k^2\right)\end{bmatrix} \tag{S14}$$

for a general many-body potential $V(q_1,\ldots,q_N)$. The N dimensional column vectors of pseudo-particle positions $\boldsymbol{q}$, $\boldsymbol{Q}$, $\boldsymbol{q}'$, $\Delta\boldsymbol{q}$, and $\overline{\boldsymbol{q}}$ are defined in the main text. The N dimensional column vector $\boldsymbol{\xi}$, whose mth element is ξ_m, is introduced by using the integral form of delta function in coarse-graining the intermediate beads. The N dimensional block column vector $\boldsymbol{a}$ has $\boldsymbol{a}_m$ as its mth block element, and the kth element of the infinite dimensional column vector $\boldsymbol{a}_m$ is $a_{m,k}$. Lastly, the N dimensional matrix $\boldsymbol{\Sigma}$ is defined in Eq. (S9a).

As shown in Eq. (10) of the main text, we assume that there are two terms in the potential: pairwise decomposable interactions V_{ij} and many-body interactions V_z. The second term in the integral on the RHS of Eq. (S3) then can be written as

$$exp\left(-\beta \int_0^1 du\, V\big(q_1(u), \dots, q_N(u)\big)\right)$$

$$= exp\left(-\beta \sum_{\langle i,j\rangle} \int_0^1 du\, V_{ij}\big(q_i(u) - q_i(u)\big)\right) exp\left(-\beta \sum_Z \int_0^1 du\, V_z\big(\boldsymbol{q}(u)\big)\right) \quad \text{(S15)}$$

Eq. (S5) of our previous work[1] shows the Taylor expanded result of the first term on the RHS of Eq. (S15). In a similar manner, we Taylor expand the many-body term. We can write

$$\int_0^1 du\, V_z\big(\boldsymbol{q}(u)\big) = \int_0^1 du\, V_z\big(\boldsymbol{q}(0) + (\boldsymbol{q}(1) - \boldsymbol{q}(0))u\big)$$

$$+ \int_0^1 du\, \boldsymbol{q}_Q(u) \cdot (DV_z|_c) + \frac{1}{2}\int_0^1 du\, \boldsymbol{q}_Q^T(u)(D^2V_z|_c)\boldsymbol{q}_Q(u) + \int_0^1 du\, \mathcal{O}(D^3V_z) \quad \text{(S16)}$$

where $DV_z|_c$ is the transposed Jacobian matrix of V_z evaluated at the classical path and $D^2V_z|_c$ is the Hessian matrix of V_z evaluated at the classical path.

For simplicity, we define a *N* dimensional column vector $\boldsymbol{D}_{Z,c}^{(1)}$ and a *N* dimensional square matrix $\boldsymbol{D}_{Z,c}^{(2)}$ such that $DV_z|_c = \boldsymbol{D}_{Z,c}^{(1)}$ and $D^2V_z|_c = \boldsymbol{D}_{Z,c}^{(2)}$. The *i*th element of $\boldsymbol{D}_{Z,c}^{(1)}$ is noted as $\left(\boldsymbol{D}_{Z,c}^{(1)}\right)_i$ and the *i*th row and *j*th column element of $\boldsymbol{D}_{Z,c}^{(2)}$ is noted as $\left(\boldsymbol{D}_{Z,c}^{(2)}\right)_{ij}$. Using this notation, integrands of the second and third term on RHS of Eq. (S16) can be written as

$$\boldsymbol{q}_Q(u) \cdot (DV_z|_c) = \sum_{i=1}^N \frac{\sqrt{2}\sigma_i}{\pi}\left(\sum_{n=1}^\infty \frac{a_{i,n}\sin(n\pi u)}{n}\right)\left(\boldsymbol{D}_{Z,c}^{(1)}\right)_i \quad \text{(S17a)}$$

$$\boldsymbol{q}_Q^T(u)(D^2V_z|_c)\boldsymbol{q}_Q(u)$$

$$= \sum_{i=1}^N \sum_{j=1}^N \frac{2\sigma_i\sigma_j}{\pi^2}\left(\sum_{n=1}^\infty \sum_{m=1}^\infty \frac{a_{i,n}a_{j,m}\sin(n\pi u)\sin(m\pi u)}{nm}\right)\left(\boldsymbol{D}_{Z,c}^{(2)}\right)_{ij}. \quad \text{(S17b)}$$

Before evaluating and simplifying Eq. (S16), we define a *N* dimensional block vector with infinite dimensional vector as the block element $\boldsymbol{b}_Z$ and a *N* dimensional square block matrix with infinite dimensional square matrix as the block element $\boldsymbol{A}_Z$. The *n*th element of the *i*th block element of $\boldsymbol{b}_Z$ and the *n*th row and *m*th column element of the *i*th row and *j*th column element of $\boldsymbol{A}_Z$ can be written as

$$(\boldsymbol{b}_Z)_{i,n} = -\beta \frac{\sqrt{2}\sigma_i}{n\pi} \int_0^1 du \left(\boldsymbol{D}_{Z,c}^{(1)}\right)_i \sin(n\pi u) \tag{S18a}$$

$$(\boldsymbol{A}_Z)_{ij,nm} = \beta \frac{2\sigma_i\sigma_j}{nm\pi^2} \int_0^1 du \left(\boldsymbol{D}_{Z,c}^{(2)}\right)_{ij} \sin(n\pi u)\sin(m\pi u). \tag{S18b}$$

Lastly, we define a vector $\boldsymbol{B}_2$ and a matrix $\boldsymbol{\Omega}_2$ with same dimensionality as $\boldsymbol{b}_Z$ and $\boldsymbol{A}_Z$ such that $\boldsymbol{B}_2 = \sum_Z \boldsymbol{b}_Z$ and $\boldsymbol{\Omega}_2 = \sum_Z \boldsymbol{A}_Z$. Note that $\boldsymbol{b}_Z$ and $\boldsymbol{A}_Z$ are two quantities for a signle many-body potential V_z and $\boldsymbol{B}_2$ and $\boldsymbol{\Omega}_2$ sum over all many-body interactions present in the force field.

Using Eqs. (S17a) and (S17b) along with the newly defined vectors, we can evaluate the integral with respect to u for the many-body potential V_Z. Eq. (S16) can be rewritten as

$$\int_0^1 du\, V_z(\boldsymbol{q}(u)) = \int_0^1 du\, V_z\big(\boldsymbol{q}(0) + (\boldsymbol{q}(1) - \boldsymbol{q}(0))u\big)$$

$$-\frac{1}{\beta}\boldsymbol{a}\cdot\boldsymbol{b}_Z + \frac{1}{2\beta}\boldsymbol{a}^T\boldsymbol{A}_Z\boldsymbol{a} + \int_0^1 du\, \mathcal{O}(D^3V_z). \tag{S19}$$

Using both Eq. (S19) and Eq. (S16) from our previous derivation[1] to rewrite Eq. (S14), we have

$$e^{-\beta V_{CG,N,trunc}} = \left(\prod_{i=1}^N \frac{\sigma_i}{\sqrt{8\pi}}\right) exp\left(-\frac{1}{2}\Delta\boldsymbol{q}^T\boldsymbol{\Sigma}\Delta\boldsymbol{q}\right)$$

$$\times\, exp\left(-\beta\int_0^1 du\, V\big(\boldsymbol{q}(0) + (\boldsymbol{q}(1) - \boldsymbol{q}(0))u\big)\right)$$

$$\times \int \frac{d\boldsymbol{\xi}d\boldsymbol{a}}{\sqrt{2\pi}} exp\left[-\frac{1}{2}\boldsymbol{a}^T(\boldsymbol{\Omega}_1 + \boldsymbol{\Omega}_2 + \boldsymbol{I})\boldsymbol{a} + (\boldsymbol{B}_1 + \boldsymbol{B}_2)\cdot\boldsymbol{a} + i\boldsymbol{\xi}\cdot(\boldsymbol{Q} - \overline{\boldsymbol{q}})\right]. \tag{S20}$$

Note that third and higher order Taylor expansion terms are not included in Eq. (S20). We name the second order truncated expression of the many-body CG potential as $V_{CG,N,trunc}$. In Eq. (S20), the matrix $\boldsymbol{\Omega}_1$ has the same dimensionality as $\boldsymbol{\Omega}_2$ and is the matrix defined in Eq. (S10) from the previous derivation, and the vector $\boldsymbol{B}_1$ has the same dimensionality as $\boldsymbol{B}_2$ as the vector defined in Eq. (S16) from the previous derivation as well.[1] Nomenclature of both quantities were changed for notational convenience.

The Gaussian integral with respect to $\boldsymbol{a}$ in Eq. (S20) can be evaluated as

$$\int \frac{d\boldsymbol{a}}{\sqrt{2\pi}} exp\left[-\frac{1}{2}\boldsymbol{a}^T(\boldsymbol{\Omega}_1 + \boldsymbol{\Omega}_2 + \boldsymbol{I})\boldsymbol{a} + (\boldsymbol{B} + \boldsymbol{B}_2)\cdot\boldsymbol{a}\right]$$

$$= \sqrt{\frac{1}{det(\mathbf{\Omega}_1+\mathbf{\Omega}_2+\boldsymbol{I})}} exp\left(\frac{1}{2}(\boldsymbol{B}+\boldsymbol{B}_2)^T\boldsymbol{\Lambda}(\boldsymbol{B}+\boldsymbol{B}_2)\right) \tag{S21}$$

for $\boldsymbol{\Lambda} = (\mathbf{\Omega}_1 + \mathbf{\Omega}_2 + \boldsymbol{I})^{-1}$. Eq. (S20) can then be simplified as

$$e^{-\beta V_{CG,N,trunc}} = \left(\prod_{i=1}^{N} \frac{\sigma_i}{\sqrt{8\pi}}\right) exp\left(-\frac{1}{2}\Delta\boldsymbol{q}^T\mathbf{\Sigma}\Delta\boldsymbol{q}\right)$$

$$\times\, exp\left(-\beta\int_0^1 du\, V\big(\boldsymbol{q}(0) + \big(\boldsymbol{q}(1)-\boldsymbol{q}(0)\big)u\big)\right)$$

$$\times \sqrt{\frac{1}{det(\mathbf{\Omega}_1+\mathbf{\Omega}_2+\boldsymbol{I})}} \int d\boldsymbol{\xi}\, exp\left[\frac{1}{2}(\boldsymbol{B}_1+\boldsymbol{B}_2)^T\boldsymbol{\Lambda}(\boldsymbol{B}_1+\boldsymbol{B}_2) + i\boldsymbol{\xi}\cdot(\boldsymbol{Q}-\overline{\boldsymbol{q}})\right]. \tag{S22}$$

Before we evaluate the integral with respect to $\boldsymbol{\xi}$ in Eq. (S22), we define matrices to simplify the notation. For the two N dimensional square matrices $\boldsymbol{L}$ and $\boldsymbol{R}$ defined in Eqs. (S8a) and (S8b), we define two N dimensional column vector $\boldsymbol{U}$ and $\boldsymbol{D}$ whose mth element are written as

$$\boldsymbol{U}_m = \sigma_m \sum_{n=1}^{N} \boldsymbol{b}_\xi{}^T\boldsymbol{\Lambda}_{mn}\boldsymbol{B}_2 \tag{S23a}$$

$$\boldsymbol{D}_m = \sigma_m \sum_{n=1}^{N} \boldsymbol{b}_\lambda{}^T\boldsymbol{\Lambda}_{mn}\boldsymbol{B}_2. \tag{S23b}$$

Using the new notation, we can expand the exponent in Eq. (S22) to have

$$\frac{1}{2}(\boldsymbol{B}_1+\boldsymbol{B}_2)^T\boldsymbol{\Lambda}(\boldsymbol{B}_1+\boldsymbol{B}_2) + i\boldsymbol{\xi}\cdot(\boldsymbol{Q}-\overline{\boldsymbol{q}})$$

$$= -\frac{1}{2}\boldsymbol{\xi}^T\boldsymbol{L}\boldsymbol{\xi} + i\boldsymbol{\xi}\left(\frac{\beta}{2}\boldsymbol{S} + \boldsymbol{U} + (\boldsymbol{Q}-\overline{\boldsymbol{q}})\right)$$

$$+\frac{\beta^2}{8}(\boldsymbol{F}^T\boldsymbol{L}\boldsymbol{F} + \boldsymbol{G}^T\boldsymbol{R}\boldsymbol{G}) + \frac{\beta}{2}(\boldsymbol{F}\cdot\boldsymbol{U} + \boldsymbol{G}\cdot\boldsymbol{D}) + \frac{1}{2}\boldsymbol{B}_2{}^T\boldsymbol{\Lambda}\boldsymbol{B}_2 \tag{S24}$$

where the two N dimensional square matrices $\boldsymbol{F}$ and $\boldsymbol{G}$ are defined according to Eqs. (S13a) and (S13b) of our previous derivation.[1] and the N dimensional column vector $\boldsymbol{S}$ satisfies $\boldsymbol{S} = \boldsymbol{L}\boldsymbol{F}$. Combine Eqs. (S22) and (S24) to have

$$e^{-\beta V_{CG,N,trunc}} = \left(\prod_{i=1}^{N} \frac{\sigma_i}{\sqrt{8\pi}}\right) exp\left(-\frac{1}{2}\Delta\boldsymbol{q}^T\mathbf{\Sigma}\Delta\boldsymbol{q}\right)$$

$$\times\, exp\left(-\beta\int_0^1 du\, V\big(\boldsymbol{q}(0) + \big(\boldsymbol{q}(1)-\boldsymbol{q}(0)\big)u\big)\right)$$

$$\times \sqrt{\frac{1}{det(\boldsymbol{\Omega}_1+\boldsymbol{\Omega}_2+\boldsymbol{I})}}\, exp\left(\frac{\beta^2}{8}(\boldsymbol{F}^T\boldsymbol{L}\boldsymbol{F}+\boldsymbol{G}^T\boldsymbol{R}\boldsymbol{G})+\frac{\beta}{2}(\boldsymbol{F}\cdot\boldsymbol{U}+\boldsymbol{G}\cdot\boldsymbol{D})+\frac{1}{2}\boldsymbol{B}_2{}^T\boldsymbol{\Lambda}\boldsymbol{B}_2\right)$$

$$\times \int d\boldsymbol{\xi}\, exp\left[-\frac{1}{2}\boldsymbol{\xi}^T\boldsymbol{L}\boldsymbol{\xi}+i\boldsymbol{\xi}\left(\frac{\beta}{2}\boldsymbol{S}+\boldsymbol{U}+(\boldsymbol{Q}-\overline{\boldsymbol{q}})\right)\right]. \tag{S25}$$

Evaluating the Gaussian integral in Eq. (S25) gives

$$e^{-\beta V_{CG,N,trunc}} = \left(\prod_{i=1}^{N}\frac{\sigma_i}{\sqrt{8\pi}}\right) exp\left(-\frac{1}{2}\Delta\boldsymbol{q}^T\boldsymbol{\Sigma}\Delta\boldsymbol{q}\right)$$

$$\times\, exp\left(-\beta\int_0^1 du\, V\big(\boldsymbol{q}(0)+(\boldsymbol{q}(1)-\boldsymbol{q}(0))u\big)\right)\sqrt{\frac{(2\pi)^N}{det(\boldsymbol{\Omega}_1+\boldsymbol{\Omega}_2+\boldsymbol{I})det(\boldsymbol{L})}}$$

$$\times\, exp\begin{pmatrix}\frac{\beta^2}{8}\boldsymbol{G}^T\boldsymbol{R}\boldsymbol{G}+\frac{\beta}{2}\boldsymbol{G}\cdot\boldsymbol{D}+\frac{1}{2}\boldsymbol{B}_2{}^T\boldsymbol{\Lambda}\boldsymbol{B}_2-\frac{1}{2}(\boldsymbol{Q}-\overline{\boldsymbol{q}})^T\boldsymbol{L}^{-1}(\boldsymbol{Q}-\overline{\boldsymbol{q}})\\ -\frac{1}{2}\boldsymbol{U}^T\boldsymbol{L}^{-1}\boldsymbol{U}-\frac{\beta}{2}\boldsymbol{F}\cdot(\boldsymbol{Q}-\overline{\boldsymbol{q}})-\boldsymbol{U}^T\boldsymbol{L}^{-1}(\boldsymbol{Q}-\overline{\boldsymbol{q}})\end{pmatrix}. \tag{S26}$$

Lastly, we can evaluate the integral with respect to u in Eq. (S18a) by Taylor expanding the classical with respect to $\overline{\boldsymbol{q}}$ and retain up to second derivative (Hessian) terms such that

$$(\boldsymbol{b}_Z)_{i,n} = -\beta\frac{\sqrt{2}\sigma_i}{n\pi}\int_0^1 du\left(\boldsymbol{D}_{Z,c}^{(1)}\right)_i \sin(n\pi u)$$

$$\cong \beta\sigma_i\left(b_{\xi,n}\left(\boldsymbol{D}_{Z,\overline{\boldsymbol{q}}}^{(1)}\right)_i + b_{\lambda,n}\left(\boldsymbol{D}_{Z,\overline{\boldsymbol{q}}}^{(2)}\right)_i\cdot\Delta\boldsymbol{q}\right) \tag{S27}$$

The term $\left(\boldsymbol{D}_{Z,\overline{\boldsymbol{q}}}^{(1)}\right)_i$ is the ith element of the transposed Jacobian matrix of V_Z evaluated at the position $\overline{\boldsymbol{q}}$. Similarly, the term $\left(\boldsymbol{D}_{Z,\overline{\boldsymbol{q}}}^{(2)}\right)_i$ is the ith row of the Hessian matrix of V_Z evaluated at the position $\overline{\boldsymbol{q}}$. Using the Taylor expanded result in Eq. (S27), we can rewrite the ith block element of $\boldsymbol{B}_2$ such that

$$(\boldsymbol{B}_2)_i = \beta\sigma_i\left(\mathcal{E}_i^{(1)}\boldsymbol{b}_\xi+\mathcal{E}_i^{(2)}\boldsymbol{b}_\lambda\right) \tag{S28}$$

for two N dimensional column vectors $\boldsymbol{\mathcal{E}}^{(1)}$ and $\boldsymbol{\mathcal{E}}^{(2)}$ whose ith elements $\mathcal{E}_i^{(1)}$ and $\mathcal{E}_i^{(2)}$ are written as

$$\mathcal{E}_i^{(1)} = \sum_Z\left(\boldsymbol{D}_{Z,\overline{\boldsymbol{q}}}^{(1)}\right)_i \tag{S29a}$$

$$\mathcal{E}_i^{(2)} = \sum_Z \left(\boldsymbol{D}_{Z,\overline{\boldsymbol{q}}}^{(2)}\right)_i \cdot \Delta\boldsymbol{q}. \tag{S29b}$$

Using this notation, we can rewrite the two vectors $\boldsymbol{U}$ and $\boldsymbol{D}$ as well such that

$$\boldsymbol{U} = \beta\boldsymbol{L}\boldsymbol{\mathcal{E}}^{(1)} \tag{S30a}$$

$$\boldsymbol{D} = \beta\boldsymbol{R}\boldsymbol{\mathcal{E}}^{(2)}. \tag{S30b}$$

Using the new notations, we can simplify Eq. (S26) to have

$$e^{-\beta V_{CG,N,trunc}} = \left(\prod_{i=1}^{N} \frac{\sigma_i}{\sqrt{8\pi}}\right) exp\left(-\frac{1}{2}\Delta\boldsymbol{q}^T\boldsymbol{\Sigma}\Delta\boldsymbol{q}\right)$$

$$\times\, exp\left(-\beta\int_0^1 du\, V\big(\boldsymbol{q}(0) + (\boldsymbol{q}(1) - \boldsymbol{q}(0))u\big)\right)\sqrt{\frac{(2\pi)^N}{det(\boldsymbol{\Omega}_1+\boldsymbol{\Omega}_2+\boldsymbol{I})det(\boldsymbol{L})}}$$

$$\times\, exp\begin{pmatrix} \frac{\beta^2}{8}\left(\boldsymbol{G} + 2\boldsymbol{\mathcal{E}}^{(2)}\right)^T\boldsymbol{R}\left(\boldsymbol{G} + 2\boldsymbol{\mathcal{E}}^{(2)}\right) \\ -\frac{1}{2}(\boldsymbol{Q} - \overline{\boldsymbol{q}})^T\boldsymbol{L}^{-1}(\boldsymbol{Q} - \overline{\boldsymbol{q}}) \\ -\frac{\beta}{2}\left(\boldsymbol{F} + 2\boldsymbol{\mathcal{E}}^{(1)}\right)\cdot(\boldsymbol{Q} - \overline{\boldsymbol{q}}) \end{pmatrix}. \tag{S31}$$

Or alternatively, the truncated many-body CG potential $V_{CG,N}$ can be written as

$$V_{CG,N,trunc} = \frac{1}{2\beta}\Delta\boldsymbol{q}^T\boldsymbol{\Sigma}\Delta\boldsymbol{q} + \int_0^1 du\, V\big(\boldsymbol{q}(0) + (\boldsymbol{q}(1) - \boldsymbol{q}(0))u\big)$$

$$-\frac{\beta}{8}\left(\boldsymbol{G} + 2\boldsymbol{\mathcal{E}}^{(2)}\right)^T\boldsymbol{R}\left(\boldsymbol{G} + 2\boldsymbol{\mathcal{E}}^{(2)}\right) + \frac{1}{2\beta}(\boldsymbol{Q} - \overline{\boldsymbol{q}})^T\boldsymbol{L}^{-1}(\boldsymbol{Q} - \overline{\boldsymbol{q}})$$

$$+\frac{1}{2}\left(\boldsymbol{F} + 2\boldsymbol{\mathcal{E}}^{(1)}\right)\cdot(\boldsymbol{Q} - \overline{\boldsymbol{q}}) - \frac{1}{\beta}\ln\left[\left(\prod_{i=1}^{N}\frac{\sigma_i}{\sqrt{8\pi}}\right)\sqrt{\frac{(2\pi)^N}{det(\boldsymbol{\Omega}_1+\boldsymbol{\Omega}_2+\boldsymbol{I})det(\boldsymbol{L})}}\right] \tag{S32}$$

The integral term in Eq. (S32) can be decomposed and Taylor expanded to have

$$\int_0^1 du\, V_{ij}\big(A_{ij} + (B_{ij} - A_{ij})u\big)$$

$$= V_{ij}\left(\overline{q}_i - \overline{q}_j\right) + \frac{1}{24}V_{ij}''\left(\overline{q}_i - \overline{q}_j\right)\cdot\left(\Delta q_i - \Delta q_j\right)^2 + \mathcal{O}\left(V_{ij}'''\right) \tag{S33}$$

and

$$\int_0^1 du\, V_Z\big(\boldsymbol{q}(0) + (\boldsymbol{q}(1) - \boldsymbol{q}(0))u\big) = V_Z(\overline{\boldsymbol{q}}) + \frac{1}{24}\Delta\boldsymbol{q}^T\boldsymbol{D}_{Z,\overline{\boldsymbol{q}}}^{(2)}\Delta\boldsymbol{q} + \mathcal{O}(D^3) \tag{S34}$$

where $A_{ij} = q_i(0) - q_j(0)$ and $B_{ij} = q_i(1) - q_j(1)$ and $\boldsymbol{D}_{Z,\overline{\boldsymbol{q}}}^{(2)}$ is the Hessian of V_Z evaluated at $\overline{\boldsymbol{q}}$.

Omitting the third and higher order terms in Eqs. (S33) and (S34), we can rewrite Eq. (S32) as

$$V_{CG,N,trunc} = \frac{1}{2\beta}\Delta\boldsymbol{q}^T\boldsymbol{\Sigma}\Delta\boldsymbol{q} + V(\overline{\boldsymbol{q}}) + \frac{1}{24}\sum_{\langle i,j\rangle} V_{ij}''\left(\overline{q}_i - \overline{q}_j\right)\cdot\left(\Delta q_i - \Delta q_j\right)^2 +$$

$$\frac{1}{24}\sum_{\boldsymbol{Z}}\Delta\boldsymbol{q}^T\boldsymbol{D}_{Z,\overline{\boldsymbol{q}}}^{(2)}\Delta\boldsymbol{q} - \frac{\beta}{8}\left(\boldsymbol{G} + 2\boldsymbol{\mathcal{E}}^{(2)}\right)^T\boldsymbol{R}\left(\boldsymbol{G} + 2\boldsymbol{\mathcal{E}}^{(2)}\right) + \frac{1}{2\beta}(\boldsymbol{Q} - \overline{\boldsymbol{q}})^T\boldsymbol{L}^{-\boldsymbol{1}}(\boldsymbol{Q} - \overline{\boldsymbol{q}}) +$$

$$\frac{1}{2}\left(\boldsymbol{F} + 2\boldsymbol{\mathcal{E}}^{(1)}\right)\cdot(\boldsymbol{Q} - \overline{\boldsymbol{q}}) - \frac{1}{\beta}\ln\left[\left(\prod_{i=1}^{N}\frac{\sigma_i}{\sqrt{8\pi}}\right)\sqrt{\frac{(2\pi)^N}{det(\boldsymbol{\Omega}_1+\boldsymbol{\Omega}_2+\boldsymbol{I})det(\boldsymbol{L})}}\right]. \quad \text{(S35)}$$

Lastly, we re-express the $V(\overline{\boldsymbol{q}})$ term by expanding $V(\boldsymbol{Q})$ at $\overline{\boldsymbol{q}}$ by making the two substitutions shown as

$$\sum_{\langle i,j\rangle} V_{ij}\left(\overline{q}_i - \overline{q}_j\right) + \frac{1}{2}\boldsymbol{F}\cdot(\boldsymbol{Q} - \overline{\boldsymbol{q}})$$

$$\rightarrow \sum_{\langle i,j\rangle} V_{ij}\left(Q_i - Q_j\right) - \frac{1}{2}V_{ij}''\left(\overline{q}_i - \overline{q}_j\right)\left(\left(Q_i - \overline{q}_i\right) - \left(Q_j - \overline{q}_j\right)\right)^2 \quad \text{(S36)}$$

and

$$\sum_{\boldsymbol{Z}} V_Z(\overline{\boldsymbol{q}}) + \boldsymbol{\mathcal{E}}^{(1)}\cdot(\boldsymbol{Q} - \overline{\boldsymbol{q}}) \rightarrow \sum_{\boldsymbol{Z}} V_Z(\boldsymbol{Q}) - \frac{1}{2}(\boldsymbol{Q} - \overline{\boldsymbol{q}})^T\boldsymbol{D}_{Z,\overline{\boldsymbol{q}}}^{(2)}(\boldsymbol{Q} - \overline{\boldsymbol{q}}). \quad \text{(S37)}$$

The LHS of Eqs. (S36) and (S37) are already included in Eq. (S35). Combining the substitution, the second order truncated Taylor expanded expression of the many-body CG potential can be written as

$$V_{CG,N,trunc} = V(\boldsymbol{Q}) + \frac{1}{2}(\boldsymbol{Q} - \overline{\boldsymbol{q}})^T\boldsymbol{K}_{Q\bar{q}}(\boldsymbol{Q} - \overline{\boldsymbol{q}}) + \frac{1}{2}\Delta\boldsymbol{q}^T\boldsymbol{K}_{\Delta q}\Delta\boldsymbol{q} + K_0 \quad \text{(S38)}$$

where

$$\boldsymbol{K}_{Q\bar{q}} = \frac{1}{\beta}\boldsymbol{L}^{-1} - \frac{1}{2}M - \sum_{\boldsymbol{Z}}\boldsymbol{D}_{Z,\overline{\boldsymbol{q}}}^{(2)} \quad \text{(S39a)}$$

$$\boldsymbol{K}_{\Delta q} = \frac{1}{24}M + \frac{1}{\beta}\boldsymbol{\Sigma} - \frac{\beta}{4}\boldsymbol{C} + \frac{1}{12}\sum_{\boldsymbol{Z}}\boldsymbol{D}_{Z,\overline{\boldsymbol{q}}}^{(2)} \quad \text{(S39b)}$$

$$K_0 = \frac{1}{2\beta}\ln\left[\left(\prod_{i=1}^{N}\frac{4m_i}{\hbar^2\beta}\right)det(\boldsymbol{\Omega}_1 + \boldsymbol{\Omega}_2 + \boldsymbol{I})det(\boldsymbol{L})\right] \quad \text{(S39c)}$$

for the N dimensional square matrices $\boldsymbol{\Sigma}$ defined in Eq. (S9a), M defined in Eq. (S9b), and $\boldsymbol{C}$ defined in Eq. (S10).

SUPPLEMENTAL REFERENCES

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