Electronic Supplementary Information for

The micromechanical model to computationally investigate cooperative and correlated phenomena in metal-organic frameworks

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S1 Derivation of the equations of motion for the micromechanical model consisting of unistable nanocells

In this section, the equations of motion for a micromechanical model that consists solely of nanocells with a single stable phase will be derived. To this end, the relation between the nanocells and the nodes of the micromechanical model will be made explicit in Section S1.1, directly leading to the Hamiltonian formulation of the micromechanical model in Section S1.2. Based on this Hamiltonian and the explicit derivatives determined in Section S1.3, the equations of motion of Section S1.4 are obtained. Finally, in Section S1.5, the extension towards nonperiodic or isolated micromechanical models is discussed.

S1.1 Definition of the relation between the nanocells and the nodes of the micromechanical model

The central idea behind the micromechanical model is to partition the extended material into nanocells $\mathbf{h}_{\mu\nu\kappa}$. As these nanocells are substantially smaller than the extended material, the 3×3 equilibrium cell matrix $\mathbf{h}_{\mu\nu\kappa,0}$ and the fourth-order 3×3×3×3 stiffness tensor $\mathbf{C}_{\mu\nu\kappa,0}$ for each of the nanocells can be determined from simulations at a higher level or theory or using a less sparse representation. This *a priori* input allows us to define at any instant during our simulation the 3×3 finite Lagrangian strain tensor $\varepsilon_{\mu\nu\kappa}$ experienced by the nanocell $\mathbf{h}_{\mu\nu\kappa}$ as

$$\varepsilon_{\mu\nu\kappa} = \frac{1}{2}\left[\mathbf{h}_{\mu\nu\kappa,0}^{-T}\mathbf{h}_{\mu\nu\kappa}^{T}\mathbf{h}_{\mu\nu\kappa}\mathbf{h}_{\mu\nu\kappa,0}^{-1} - \mathbf{1}\right], \tag{S1.1}$$

in which $\mathbf{1}$ is the 3×3 unit tensor. In turn, this leads automatically to the following potential energy associated with each of the nanocells:

$$\mathcal{U}_{\mu\nu\kappa} = \frac{1}{2}\det(\mathbf{h}_{\mu\nu\kappa})\varepsilon_{\mu\nu\kappa}^{T} : \mathbf{C}_{\mu\nu\kappa,0} : \varepsilon_{\mu\nu\kappa}. \tag{S1.2}$$

In this expression, ':' denotes the matrix inproduct:

$$\left[\varepsilon^{T} : \mathbf{C}\right]_{k\ell} = \sum_{i=1}^{3}\sum_{j=1}^{3}\varepsilon_{ji}C_{ijk\ell} \quad \text{and} \quad [\mathbf{C} : \varepsilon]_{k\ell} = \sum_{i=1}^{3}\sum_{j=1}^{3}C_{k\ell ij}\varepsilon_{ij}. \tag{S1.3}$$

Each nanocell $\mathbf{h}_{\mu\nu\kappa}$ in our partitioning is defined by the vectors connecting the eight nodes on its corners, as indicated in Figure S1 for an arbitrary nanocell. Using the notation of Figure S1 and with $\boldsymbol{r}_{ijk}$ the position of node N_{ijk}, we thus find:

$$\mathbf{h}_{000} = \frac{1}{4}\begin{bmatrix} (\boldsymbol{r}_{100}^T - \boldsymbol{r}_{000}^T) + (\boldsymbol{r}_{110}^T - \boldsymbol{r}_{010}^T) + (\boldsymbol{r}_{101}^T - \boldsymbol{r}_{001}^T) + (\boldsymbol{r}_{111}^T - \boldsymbol{r}_{011}^T) \\ (\boldsymbol{r}_{010}^T - \boldsymbol{r}_{000}^T) + (\boldsymbol{r}_{110}^T - \boldsymbol{r}_{100}^T) + (\boldsymbol{r}_{011}^T - \boldsymbol{r}_{001}^T) + (\boldsymbol{r}_{111}^T - \boldsymbol{r}_{101}^T) \\ (\boldsymbol{r}_{001}^T - \boldsymbol{r}_{000}^T) + (\boldsymbol{r}_{011}^T - \boldsymbol{r}_{010}^T) + (\boldsymbol{r}_{101}^T - \boldsymbol{r}_{100}^T) + (\boldsymbol{r}_{111}^T - \boldsymbol{r}_{110}^T) \end{bmatrix}. \tag{S1.4}$$

In this notation, the indices (μ, ν, κ) and (i, j, k) were shifted so that $\mu = \nu = \kappa = 0$. To obtain the expression for nanocell $\mathbf{h}_{\mu\nu\kappa}$, it therefore suffices to add μ, ν, and κ to the i, j and k indices of the position vectors, respectively.

S1.2 Hamiltonian formulation of the micromechanical model

Let $\boldsymbol{r} = \{\boldsymbol{r}_{ijk}\}$ be the set of position vectors of the nodes in the micromechanical model, with $i \in [0, n_x - 1]$, $j \in [0, n_y - 1]$, and $k \in [0, n_z - 1]$, and let $\dot{\boldsymbol{r}} = \{\dot{\boldsymbol{r}}_{ijk}\}$ be the set of velocity vectors of the nodes. At this moment, periodic boundary conditions are assumed, although the extension to isolated systems can be made easily (see Section S1.5). As a result of these periodic boundary

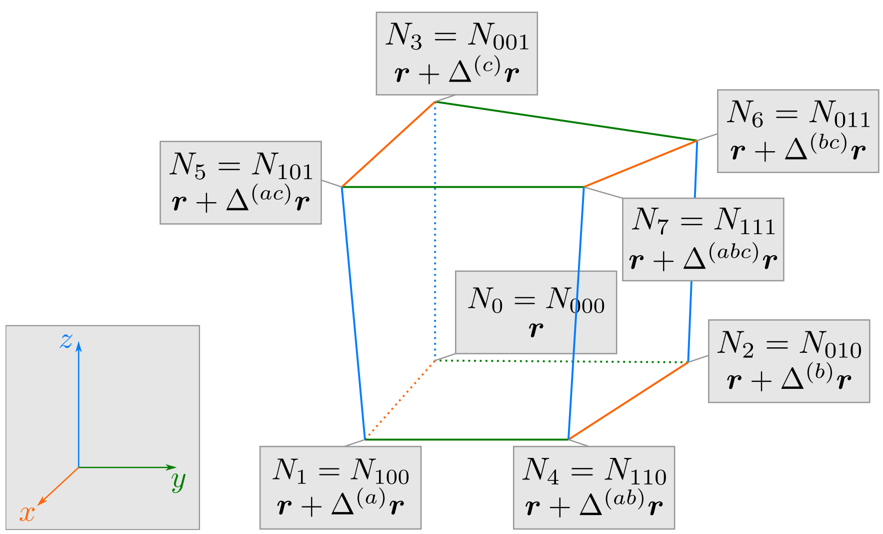

Figure S1: Representation of an arbitrary nanocell $\mathbf{h}_{\mu\nu\kappa}$ and the eight nodes N_{ijk} that define the cell. For notational simplicity, the indexing has been shifted so that $\mu = \nu = \kappa = 0$ in this representation. This means that for an arbitrary nanocell, the nodes will be given by $N_0 = N_{i=\mu,j=\nu,k=\kappa}$, $N_1 = N_{i=\mu+1,j=\nu,k=\kappa}$, ..., until $N_7 = N_{i=\mu+1,j=\nu+1,k=\kappa+1}$.

conditions, $\boldsymbol{r}_{n_x jk} = \boldsymbol{r}_{0jk}, \forall j, k$, and similar expressions hold for the other two directions. Furthermore, let m_{ijk} be the mass associated with the node N_{ijk}. Typically, this mass will be chosen as the average mass of the eight nanocells for which the node N_{ijk} forms one of the corners, as indicated in Figure S2, although this is not required.

Through Eq. (S1.4), knowledge of all position vectors allows us to define all nanocells in our micromechanical model, which in turns yields the strain and elastic potential energy associated with each nanocell via Eq. (S1.1) and Eq. (S1.2). Therefore, the Lagrangian of the total microme-

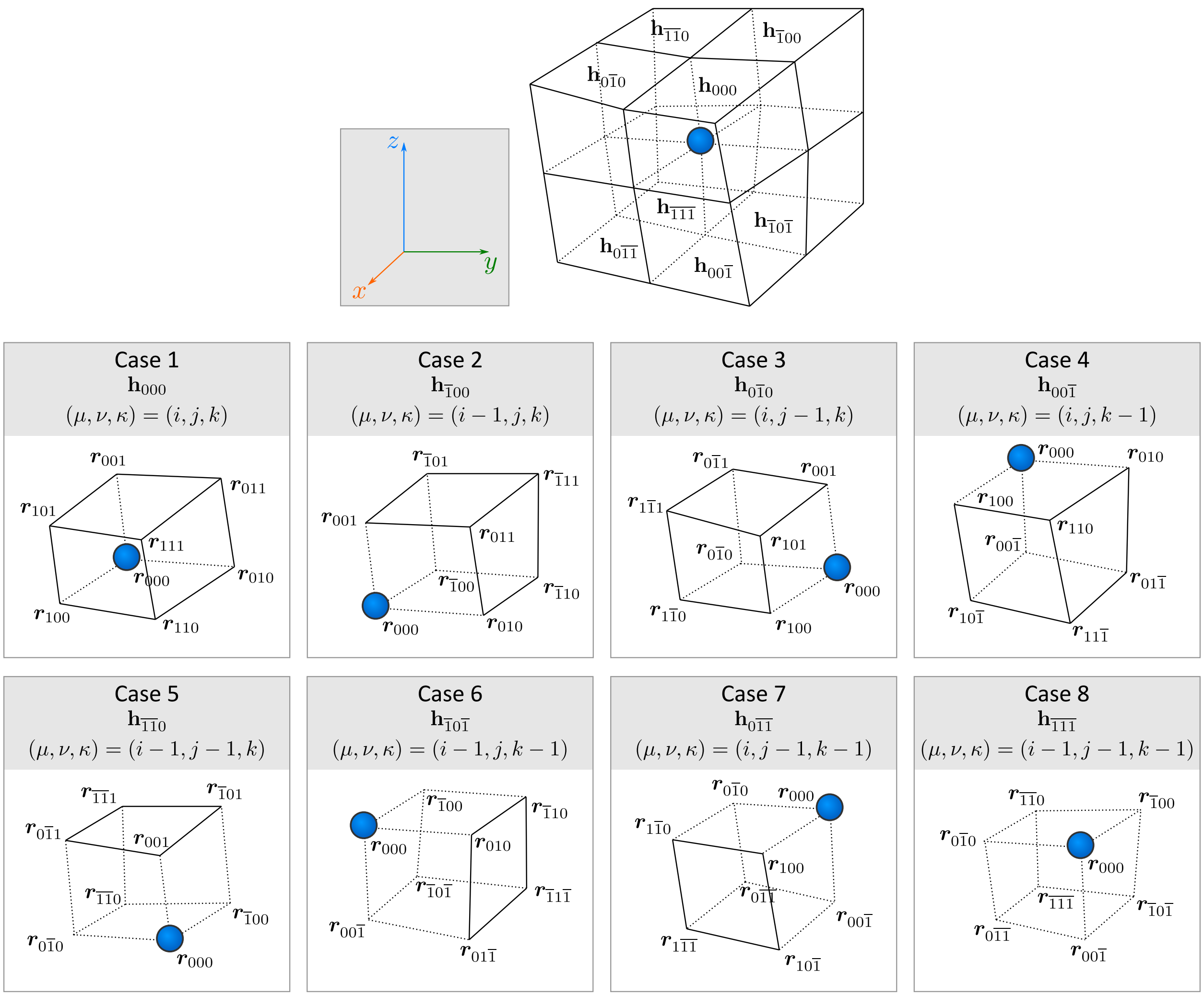

Figure S2: Overview of the eight possible nanocells $\mathbf{h}_{\mu\nu\kappa}$ that have a given node N_{ijk} (blue sphere) on one of their corners and the relation between the indices (μ, ν, κ) of these nanocells and the indices (i, j, k) of the blue node. All indices are shifted so that the blue node has indices $i = j = k = 0$; while '$\bar{1}$' is used to denote '-1'.

chanical system is given by

$$\mathcal{L}(\boldsymbol{r},\dot{\boldsymbol{r}}) = \sum_{i=0}^{n_x-1}\sum_{j=0}^{n_y-1}\sum_{k=0}^{n_z-1}\frac{1}{2}m_{ijk}\dot{\boldsymbol{r}}_{ijk}^2 - \sum_{\mu=0}^{n_x-1}\sum_{\nu=0}^{n_y-1}\sum_{\kappa=0}^{n_z-1}\mathcal{U}_{\mu\nu\kappa}(\boldsymbol{r}), \tag{S1.5}$$

with $\boldsymbol{r} = \{\boldsymbol{r}_{ijk}\}_{i=0,\ j=0,\ k=0}^{i=n_x-1,\ j=n_y-1,\ k=n_z-1}$ and $\dot{\boldsymbol{r}} = \{\dot{\boldsymbol{r}}_{ijk}\}_{i=0,\ j=0,\ k=0}^{i=n_x-1,\ j=n_y-1,\ k=n_z-1}$.

Based on this Lagrangian, the conjugated momenta can be defined as

$$\boldsymbol{p}_{ijk} = \nabla_{\dot{\boldsymbol{r}}_{ijk}}\mathcal{L}(\boldsymbol{r},\dot{\boldsymbol{r}}) = m_{ijk}\dot{\boldsymbol{r}}_{ijk} = m_{ijk}\boldsymbol{v}_{ijk}. \tag{S1.6}$$

This derivation results in the following Hamiltonian for the micromechanical model:

$$\mathcal{H}(\boldsymbol{r},\boldsymbol{p}) = \sum_{i=0}^{n_x-1}\sum_{j=0}^{n_y-1}\sum_{k=0}^{n_z-1}\frac{\boldsymbol{p}_{ijk}^2}{2m_{ijk}} + \sum_{\mu=0}^{n_x-1}\sum_{\nu=0}^{n_y-1}\sum_{\kappa=0}^{n_z-1}\mathcal{U}_{\mu\nu\kappa}(\boldsymbol{r}), \tag{S1.7}$$

with again $\boldsymbol{r} = \{\boldsymbol{r}_{ijk}\}_{i=0,\ j=0,\ k=0}^{i=n_x-1,\ j=n_y-1,\ k=n_z-1}$ and $\boldsymbol{p} = \{\boldsymbol{p}_{ijk}\}_{i=0,\ j=0,\ k=0}^{i=n_x-1,\ j=n_y-1,\ k=n_z-1}$.

From this Hamiltonian, the equations of motion can be derived as

$$\dot{\boldsymbol{r}}_{ijk} = \nabla_{\boldsymbol{p}_{ijk}}\mathcal{H} = \frac{\boldsymbol{p}_{ijk}}{m_{ijk}} = \boldsymbol{v}_{ijk}, \tag{S1.8}$$

and

$$\dot{\boldsymbol{p}}_{ijk} = -\nabla_{\boldsymbol{r}_{ijk}}\mathcal{H} = -\sum_{\mu=0}^{n_x-1}\sum_{\nu=0}^{n_y-1}\sum_{\kappa=0}^{n_z-1}\nabla_{\boldsymbol{r}_{ijk}}\mathcal{U}_{\mu\nu\kappa}(\boldsymbol{r}) = -\sum_{\mu=i-1}^{i}\sum_{\nu=j-1}^{j}\sum_{\kappa=k-1}^{k}\nabla_{\boldsymbol{r}_{ijk}}\mathcal{U}_{\mu\nu\kappa}(\boldsymbol{r}). \tag{S1.9}$$

To obtain the last expression, we used that Eq. (S1.4) implies that the position vector $\boldsymbol{r}_{ijk}$ only enters the definition of the eight nanocells in which $\mu \in \{i-1, i\}$, $\nu \in \{j-1, j\}$ and $\kappa \in \{k-1, k\}$, which is also visually depicted in Figure S2. In Section S1.3, the derivatives entering Eq. (S1.9) will be determined explicitly.

S1.3 Derivatives of the elastic potential energy

As indicated in Eq. (S1.9), updating the node's momenta requires derivatives of the elastic potential energy in Eq. (S1.2) with respect to the position vector $\boldsymbol{r}_{ijk}$:

$$\begin{aligned}
-\nabla_{\boldsymbol{r}_{ijk}}\mathcal{U}_{\mu\nu\kappa}(\boldsymbol{r}) \quad = \quad & -\frac{1}{2}\left(\nabla_{\boldsymbol{r}_{ijk}}\det(\mathbf{h}_{\mu\nu\kappa})\right)\boldsymbol{\varepsilon}^{T}_{\mu\nu\kappa} : \mathbf{C}_{\mu\nu\kappa,0} : \boldsymbol{\varepsilon}_{\mu\nu\kappa} - \frac{1}{2}\det(\mathbf{h}_{\mu\nu\kappa})\left(\nabla_{\boldsymbol{r}_{ijk}}\boldsymbol{\varepsilon}^{T}_{\mu\nu\kappa}\right) : \mathbf{C}_{\mu\nu\kappa,0} : \boldsymbol{\varepsilon}_{\mu\nu\kappa} \\
& -\frac{1}{2}\det(\mathbf{h}_{\mu\nu\kappa})\boldsymbol{\varepsilon}^{T}_{\mu\nu\kappa} : \mathbf{C}_{\mu\nu\kappa,0} : \left(\nabla_{\boldsymbol{r}_{ijk}}\boldsymbol{\varepsilon}_{\mu\nu\kappa}\right). \qquad \text{(S1.10)}
\end{aligned}$$

In this expression, we have:

$$\nabla_{\boldsymbol{r}_{ijk}}\det(\mathbf{h}_{\mu\nu\kappa}) = \det(\mathbf{h}_{\mu\nu\kappa})\mathrm{Tr}\left\{\mathbf{h}^{-1}_{\mu\nu\kappa}\nabla_{\boldsymbol{r}_{ijk}}\mathbf{h}_{\mu\nu\kappa}\right\}, \qquad \text{(S1.11)}$$

and

$$\nabla_{\boldsymbol{r}_{ijk}}\boldsymbol{\varepsilon}^{T}_{\mu\nu\kappa} = \nabla_{\boldsymbol{r}_{ijk}}\boldsymbol{\varepsilon}_{\mu\nu\kappa} = \frac{1}{2}\mathbf{h}^{-T}_{\mu\nu\kappa,0}\left(\left(\nabla_{\boldsymbol{r}_{ijk}}\mathbf{h}^{T}_{\mu\nu\kappa}\right)\mathbf{h}_{\mu\nu\kappa} + \mathbf{h}^{T}_{\mu\nu\kappa}\left(\nabla_{\boldsymbol{r}_{ijk}}\mathbf{h}_{\mu\nu\kappa}\right)\right)\mathbf{h}^{-1}_{\mu\nu\kappa,0}. \qquad \text{(S1.12)}$$

In these expressions, μ, ν, and κ are restricted to $\mu \in \{i-1, i\}$, $\nu \in \{j-1, j\}$ and $\kappa \in \{k-1, k\}$. Below, these eight cases, visualized in Figure S2, are treated separately.

Case 1: $\mathbf{h}_{\mu\nu\kappa} = \mathbf{h}_{ijk}$

In the case that $\mathbf{h}_{\mu\nu\kappa} = \mathbf{h}_{ijk}$, $\boldsymbol{r}_{000}$ in Eq. (S1.4) corresponds with $\boldsymbol{r}_{ijk}$ (recall that in Eq. (S1.4) all indices are shifted by (μ, ν, κ)). As a result:

$$\frac{\partial \mathbf{h}_{ijk}}{\partial x_{ijk}} = \frac{1}{4}\begin{bmatrix} -1 & 0 & 0 \\ -1 & 0 & 0 \\ -1 & 0 & 0 \end{bmatrix} \qquad \frac{\partial \mathbf{h}_{ijk}}{\partial y_{ijk}} = \frac{1}{4}\begin{bmatrix} 0 & -1 & 0 \\ 0 & -1 & 0 \\ 0 & -1 & 0 \end{bmatrix} \qquad \frac{\partial \mathbf{h}_{ijk}}{\partial z_{ijk}} = \frac{1}{4}\begin{bmatrix} 0 & 0 & -1 \\ 0 & 0 & -1 \\ 0 & 0 & -1 \end{bmatrix}. \qquad \text{(S1.13)}$$

As a consequence, we find for the strain derivative:

$$\frac{\partial \varepsilon_{ijk}}{\partial x_{ijk}} = -\frac{1}{8}\mathbf{h}_{ijk,0}^{-T}\left(\begin{bmatrix} 1 & 1 & 1 \\ 0 & 0 & 0 \\ 0 & 0 & 0 \end{bmatrix}\mathbf{h}_{ijk} + \mathbf{h}_{ijk}^{T}\begin{bmatrix} 1 & 0 & 0 \\ 1 & 0 & 0 \\ 1 & 0 & 0 \end{bmatrix}\right)\mathbf{h}_{ijk,0}^{-1} \tag{S1.14}$$

$$= \frac{1}{32}\mathbf{h}_{ijk,0}^{-T}\left(\begin{bmatrix} f_1(x_{ijk}) & f_1(y_{ijk}) & f_1(z_{ijk}) \\ 0 & 0 & 0 \\ 0 & 0 & 0 \end{bmatrix} + \begin{bmatrix} f_1(x_{ijk}) & 0 & 0 \\ f_1(y_{ijk}) & 0 & 0 \\ f_1(z_{ijk}) & 0 & 0 \end{bmatrix}\right)\mathbf{h}_{ijk,0}^{-1} \tag{S1.15}$$

$$= \frac{1}{32}\mathbf{h}_{ijk,0}^{-T}\begin{bmatrix} 2f_1(x_{ijk}) & f_1(y_{ijk}) & f_1(z_{ijk}) \\ f_1(y_{ijk}) & 0 & 0 \\ f_1(z_{ijk}) & 0 & 0 \end{bmatrix}\mathbf{h}_{ijk,0}^{-1}. \tag{S1.16}$$

with

$$f_1(x_{ijk}) = 3x_{ijk} + x_{i+1jk} + x_{ij+1k} + x_{ijk+1} - x_{i+1j+1k} - x_{i+1jk+1} - x_{ij+1k+1} - 3x_{i+1j+1k+1}. \tag{S1.17}$$

Similarly

$$\frac{\partial \varepsilon_{ijk}}{\partial y_{ijk}} = \frac{1}{32}\mathbf{h}_{ijk,0}^{-T}\begin{bmatrix} 0 & f_1(x_{ijk}) & 0 \\ f_1(x_{ijk}) & 2f_1(y_{ijk}) & f_1(z_{ijk}) \\ 0 & f_1(z_{ijk}) & 0 \end{bmatrix}\mathbf{h}_{ijk,0}^{-1} \tag{S1.18}$$

and

$$\frac{\partial \varepsilon_{ijk}}{\partial z_{ijk}} = \frac{1}{32}\mathbf{h}_{ijk,0}^{-T}\begin{bmatrix} 0 & 0 & f_1(x_{ijk}) \\ 0 & 0 & f_1(y_{ijk}) \\ f_1(x_{ijk}) & f_1(y_{ijk}) & 2f_1(z_{ijk}) \end{bmatrix}\mathbf{h}_{ijk,0}^{-1}. \tag{S1.19}$$

Case 2: $\mathbf{h}_{\mu\nu\kappa} = \mathbf{h}_{i-1jk}$

In the case that $\mathbf{h}_{\mu\nu\kappa} = \mathbf{h}_{i-1jk}$, $\boldsymbol{r}_{100}$ in Eq. (S1.4) corresponds with $\boldsymbol{r}_{ijk}$. As a result:

$$\frac{\partial \mathbf{h}_{i-1jk}}{\partial x_{ijk}} = \frac{1}{4}\begin{bmatrix} 1 & 0 & 0 \\ -1 & 0 & 0 \\ -1 & 0 & 0 \end{bmatrix} \qquad \frac{\partial \mathbf{h}_{i-1jk}}{\partial y_{ijk}} = \frac{1}{4}\begin{bmatrix} 0 & 1 & 0 \\ 0 & -1 & 0 \\ 0 & -1 & 0 \end{bmatrix} \qquad \frac{\partial \mathbf{h}_{i-1jk}}{\partial z_{ijk}} = \frac{1}{4}\begin{bmatrix} 0 & 0 & 1 \\ 0 & 0 & -1 \\ 0 & 0 & -1 \end{bmatrix}. \tag{S1.20}$$

As a consequence, we find for the strain derivative:

$$\frac{\partial \varepsilon_{i-1jk}}{\partial x_{ijk}} = \frac{1}{8}\mathbf{h}^{-T}_{i-1jk,0}\left(\begin{bmatrix} 1 & -1 & -1 \\ 0 & 0 & 0 \\ 0 & 0 & 0 \end{bmatrix}\mathbf{h}_{i-1jk} + \mathbf{h}^{T}_{i-1jk}\begin{bmatrix} 1 & 0 & 0 \\ -1 & 0 & 0 \\ -1 & 0 & 0 \end{bmatrix}\right)\mathbf{h}^{-1}_{i-1jk,0} \tag{S1.21}$$

$$= \frac{1}{32}\mathbf{h}^{-T}_{i-1jk,0}\left(\begin{bmatrix} f_2(x_{ijk}) & f_2(y_{ijk}) & f_2(z_{ijk}) \\ 0 & 0 & 0 \\ 0 & 0 & 0 \end{bmatrix} + \begin{bmatrix} f_2(x_{ijk}) & 0 & 0 \\ f_2(y_{ijk}) & 0 & 0 \\ f_2(z_{ijk}) & 0 & 0 \end{bmatrix}\right)\mathbf{h}^{-1}_{i-1jk,0} \tag{S1.22}$$

$$= \frac{1}{32}\mathbf{h}^{-T}_{i-1jk,0}\begin{bmatrix} 2f_2(x_{ijk}) & f_2(y_{ijk}) & f_2(z_{ijk}) \\ f_2(y_{ijk}) & 0 & 0 \\ f_2(z_{ijk}) & 0 & 0 \end{bmatrix}\mathbf{h}^{-1}_{i-1jk,0}. \tag{S1.23}$$

with

$$f_2(x_{ijk}) = x_{i-1jk} + 3x_{ijk} - x_{i-1j+1k} - x_{i-1jk+1} + x_{ij+1k} + x_{ijk+1} - 3x_{i-1j+1k+1} - x_{ij+1k+1}. \tag{S1.24}$$

Similarly

$$\frac{\partial \varepsilon_{i-1jk}}{\partial y_{ijk}} = \frac{1}{32}\mathbf{h}^{-T}_{i-1jk,0}\begin{bmatrix} 0 & f_2(x_{ijk}) & 0 \\ f_2(x_{ijk}) & 2f_2(y_{ijk}) & f_2(z_{ijk}) \\ 0 & f_2(z_{ijk}) & 0 \end{bmatrix}\mathbf{h}^{-1}_{i-1jk,0} \tag{S1.25}$$

and

$$\frac{\partial \varepsilon_{i-1jk}}{\partial z_{ijk}} = \frac{1}{32}\mathbf{h}_{i-1jk,0}^{-T}\begin{bmatrix} 0 & 0 & f_2(x_{ijk}) \\ 0 & 0 & f_2(y_{ijk}) \\ f_2(x_{ijk}) & f_2(y_{ijk}) & 2f_2(z_{ijk}) \end{bmatrix}\mathbf{h}_{i-1jk,0}^{-1}. \tag{S1.26}$$

Case 3: $\mathbf{h}_{\mu\nu\kappa} = \mathbf{h}_{ij-1k}$

In the case that $\mathbf{h}_{\mu\nu\kappa} = \mathbf{h}_{ij-1k}$, $\boldsymbol{r}_{010}$ in Eq. (S1.4) corresponds with $\boldsymbol{r}_{ijk}$. As a result:

$$\frac{\partial \mathbf{h}_{ij-1k}}{\partial x_{ijk}} = \frac{1}{4}\begin{bmatrix} -1 & 0 & 0 \\ 1 & 0 & 0 \\ -1 & 0 & 0 \end{bmatrix} \qquad \frac{\partial \mathbf{h}_{ij-1k}}{\partial y_{ijk}} = \frac{1}{4}\begin{bmatrix} 0 & -1 & 0 \\ 0 & 1 & 0 \\ 0 & -1 & 0 \end{bmatrix} \qquad \frac{\partial \mathbf{h}_{ij-1k}}{\partial z_{ijk}} = \frac{1}{4}\begin{bmatrix} 0 & 0 & -1 \\ 0 & 0 & 1 \\ 0 & 0 & -1 \end{bmatrix}. \tag{S1.27}$$

As a consequence, we find for the strain derivative:

$$\begin{aligned} \frac{\partial \varepsilon_{ij-1k}}{\partial x_{ijk}} &= \frac{1}{8}\mathbf{h}_{ij-1k,0}^{-T}\left(\begin{bmatrix} -1 & 1 & -1 \\ 0 & 0 & 0 \\ 0 & 0 & 0 \end{bmatrix}\mathbf{h}_{ij-1k} + \mathbf{h}_{ij-1k}^{T}\begin{bmatrix} -1 & 0 & 0 \\ 1 & 0 & 0 \\ -1 & 0 & 0 \end{bmatrix}\right)\mathbf{h}_{ij-1k,0}^{-1} & \text{(S1.28)} \\ &= \frac{1}{32}\mathbf{h}_{ij-1k,0}^{-T}\left(\begin{bmatrix} f_3(x_{ijk}) & f_3(y_{ijk}) & f_3(z_{ijk}) \\ 0 & 0 & 0 \\ 0 & 0 & 0 \end{bmatrix} + \begin{bmatrix} f_3(x_{ijk}) & 0 & 0 \\ f_3(y_{ijk}) & 0 & 0 \\ f_3(z_{ijk}) & 0 & 0 \end{bmatrix}\right)\mathbf{h}_{ij-1k,0}^{-1} & \text{(S1.29)} \\ &= \frac{1}{32}\mathbf{h}_{ij-1k,0}^{-T}\begin{bmatrix} 2f_3(x_{ijk}) & f_3(y_{ijk}) & f_3(z_{ijk}) \\ f_3(y_{ijk}) & 0 & 0 \\ f_3(z_{ijk}) & 0 & 0 \end{bmatrix}\mathbf{h}_{ij-1k,0}^{-1}. & \text{(S1.30)} \end{aligned}$$

with

$$f_3(x_{ijk}) = x_{ij-1k} - x_{i+1j-1k} + 3x_{ijk} - x_{ij-1k+1} + x_{i+1jk} - 3x_{i+1j-1k+1} + x_{ijk+1} - x_{i+1jk+1}. \tag{S1.31}$$

Similarly

$$\frac{\partial \varepsilon_{ij-1k}}{\partial y_{ijk}} = \frac{1}{32}\mathbf{h}_{ij-1k,0}^{-T}\begin{bmatrix} 0 & f_3(x_{ijk}) & 0 \\ f_3(x_{ijk}) & 2f_3(y_{ijk}) & f_3(z_{ijk}) \\ 0 & f_3(z_{ijk}) & 0 \end{bmatrix}\mathbf{h}_{ij-1k,0}^{-1} \tag{S1.32}$$

and

$$\frac{\partial \varepsilon_{ij-1k}}{\partial z_{ijk}} = \frac{1}{32}\mathbf{h}_{ij-1k,0}^{-T}\begin{bmatrix} 0 & 0 & f_3(x_{ijk}) \\ 0 & 0 & f_3(y_{ijk}) \\ f_3(x_{ijk}) & f_3(y_{ijk}) & 2f_3(z_{ijk}) \end{bmatrix}\mathbf{h}_{ij-1k,0}^{-1}. \tag{S1.33}$$

Case 4: $\mathbf{h}_{\mu\nu\kappa} = \mathbf{h}_{ijk-1}$

In the case that $\mathbf{h}_{\mu\nu\kappa} = \mathbf{h}_{ijk-1}$, $\boldsymbol{r}_{001}$ in Eq. (S1.4) corresponds with $\boldsymbol{r}_{ijk}$. As a result:

$$\frac{\partial \mathbf{h}_{ijk-1}}{\partial x_{ijk}} = \frac{1}{4}\begin{bmatrix} -1 & 0 & 0 \\ -1 & 0 & 0 \\ 1 & 0 & 0 \end{bmatrix} \qquad \frac{\partial \mathbf{h}_{ijk-1}}{\partial y_{ijk}} = \frac{1}{4}\begin{bmatrix} 0 & -1 & 0 \\ 0 & -1 & 0 \\ 0 & 1 & 0 \end{bmatrix} \qquad \frac{\partial \mathbf{h}_{ijk-1}}{\partial z_{ijk}} = \frac{1}{4}\begin{bmatrix} 0 & 0 & -1 \\ 0 & 0 & -1 \\ 0 & 0 & 1 \end{bmatrix}. \tag{S1.34}$$

As a consequence, we find for the strain derivative:

$$\begin{aligned}
\frac{\partial \varepsilon_{ijk-1}}{\partial x_{ijk}} &= \frac{1}{8}\mathbf{h}_{ijk-1,0}^{-T}\left(\begin{bmatrix} -1 & -1 & 1 \\ 0 & 0 & 0 \\ 0 & 0 & 0 \end{bmatrix}\mathbf{h}_{ijk-1} + \mathbf{h}_{ijk-1}^{T}\begin{bmatrix} -1 & 0 & 0 \\ -1 & 0 & 0 \\ 1 & 0 & 0 \end{bmatrix}\right)\mathbf{h}_{ijk-1,0}^{-1} && \text{(S1.35)} \\
&= \frac{1}{32}\mathbf{h}_{ijk-1,0}^{-T}\left(\begin{bmatrix} f_4(x_{ijk}) & f_4(y_{ijk}) & f_4(z_{ijk}) \\ 0 & 0 & 0 \\ 0 & 0 & 0 \end{bmatrix} + \begin{bmatrix} f_4(x_{ijk}) & 0 & 0 \\ f_4(y_{ijk}) & 0 & 0 \\ f_4(z_{ijk}) & 0 & 0 \end{bmatrix}\right)\mathbf{h}_{ijk-1,0}^{-1} && \text{(S1.36)} \\
&= \frac{1}{32}\mathbf{h}_{ijk-1,0}^{-T}\begin{bmatrix} 2f_4(x_{ijk}) & f_4(y_{ijk}) & f_4(z_{ijk}) \\ f_4(y_{ijk}) & 0 & 0 \\ f_4(z_{ijk}) & 0 & 0 \end{bmatrix}\mathbf{h}_{ijk-1,0}^{-1}. && \text{(S1.37)}
\end{aligned}$$

with

$$f_4(x_{ijk}) = x_{ijk-1} - x_{i+1jk-1} - x_{ij+1k-1} + 3x_{ijk} - 3x_{i+1j+1k-1} + x_{i+1jk} + x_{ij+1k} - x_{i+1j+1k}. \tag{S1.38}$$

Similarly

$$\frac{\partial \varepsilon_{ijk-1}}{\partial y_{ijk}} = \frac{1}{32}\mathbf{h}_{ijk-1,0}^{-T} \begin{bmatrix} 0 & f_4(x_{ijk}) & 0 \\ f_4(x_{ijk}) & 2f_4(y_{ijk}) & f_4(z_{ijk}) \\ 0 & f_4(z_{ijk}) & 0 \end{bmatrix} \mathbf{h}_{ijk-1,0}^{-1} \tag{S1.39}$$

and

$$\frac{\partial \varepsilon_{ijk-1}}{\partial z_{ijk}} = \frac{1}{32}\mathbf{h}_{ijk-1,0}^{-T} \begin{bmatrix} 0 & 0 & f_4(x_{ijk}) \\ 0 & 0 & f_4(y_{ijk}) \\ f_4(x_{ijk}) & f_4(y_{ijk}) & 2f_4(z_{ijk}) \end{bmatrix} \mathbf{h}_{ijk-1,0}^{-1}. \tag{S1.40}$$

Case 5: $\mathbf{h}_{\mu\nu\kappa} = \mathbf{h}_{i-1j-1k}$

In the case that $\mathbf{h}_{\mu\nu\kappa} = \mathbf{h}_{i-1j-1k}$, $\boldsymbol{r}_{110}$ in Eq. (S1.4) corresponds with $\boldsymbol{r}_{ijk}$. As a result:

$$\frac{\partial \mathbf{h}_{i-1j-1k}}{\partial x_{ijk}} = \frac{1}{4}\begin{bmatrix} 1 & 0 & 0 \\ 1 & 0 & 0 \\ -1 & 0 & 0 \end{bmatrix} \quad \frac{\partial \mathbf{h}_{i-1j-1k}}{\partial y_{ijk}} = \frac{1}{4}\begin{bmatrix} 0 & 1 & 0 \\ 0 & 1 & 0 \\ 0 & -1 & 0 \end{bmatrix} \quad \frac{\partial \mathbf{h}_{i-1j-1k}}{\partial z_{ijk}} = \frac{1}{4}\begin{bmatrix} 0 & 0 & 1 \\ 0 & 0 & 1 \\ 0 & 0 & -1 \end{bmatrix}. \tag{S1.41}$$

As a consequence, we find for the strain derivative:

$$\begin{aligned}
\frac{\partial \varepsilon_{i-1j-1k}}{\partial x_{ijk}} &= \frac{1}{8}\mathbf{h}^{-T}_{i-1j-1k,0}\left(\begin{bmatrix} 1 & 1 & -1 \\ 0 & 0 & 0 \\ 0 & 0 & 0 \end{bmatrix}\mathbf{h}_{i-1j-1k} + \mathbf{h}^{T}_{i-1j-1k}\begin{bmatrix} 1 & 0 & 0 \\ 1 & 0 & 0 \\ -1 & 0 & 0 \end{bmatrix}\right)\mathbf{h}^{-1}_{i-1j-1k,0} &\text{(S1.42)}\\
&= \frac{1}{32}\mathbf{h}^{-T}_{i-1j-1k,0}\left(\begin{bmatrix} f_5(x_{ijk}) & f_5(y_{ijk}) & f_5(z_{ijk}) \\ 0 & 0 & 0 \\ 0 & 0 & 0 \end{bmatrix} + \begin{bmatrix} f_5(x_{ijk}) & 0 & 0 \\ f_5(y_{ijk}) & 0 & 0 \\ f_5(z_{ijk}) & 0 & 0 \end{bmatrix}\right)\mathbf{h}^{-1}_{i-1j-1k,0} &\text{(S1.43)}\\
&= \frac{1}{32}\mathbf{h}^{-T}_{i-1j-1k,0}\begin{bmatrix} 2f_5(x_{ijk}) & f_5(y_{ijk}) & f_5(z_{ijk}) \\ f_5(y_{ijk}) & 0 & 0 \\ f_5(z_{ijk}) & 0 & 0 \end{bmatrix}\mathbf{h}^{-1}_{i-1j-1k,0}. &\text{(S1.44)}
\end{aligned}$$

with

$$f_5(x_{ijk}) = -x_{i-1j-1k} + x_{ij-1k} + x_{i-1jk} - 3x_{i-1j-1k+1} + 3x_{ijk} - x_{ij-1k+1} - x_{i-1jk+1} + x_{ijk+1}. \quad \text{(S1.45)}$$

Similarly

$$\frac{\partial \varepsilon_{i-1j-1k}}{\partial y_{ijk}} = \frac{1}{32}\mathbf{h}^{-T}_{i-1j-1k,0}\begin{bmatrix} 0 & f_5(x_{ijk}) & 0 \\ f_5(x_{ijk}) & 2f_5(y_{ijk}) & f_5(z_{ijk}) \\ 0 & f_5(z_{ijk}) & 0 \end{bmatrix}\mathbf{h}^{-1}_{i-1j-1k,0} \quad \text{(S1.46)}$$

and

$$\frac{\partial \varepsilon_{i-1j-1k}}{\partial z_{ijk}} = \frac{1}{32}\mathbf{h}^{-T}_{i-1j-1k,0}\begin{bmatrix} 0 & 0 & f_5(x_{ijk}) \\ 0 & 0 & f_5(y_{ijk}) \\ f_5(x_{ijk}) & f_5(y_{ijk}) & 2f_5(z_{ijk}) \end{bmatrix}\mathbf{h}^{-1}_{i-1j-1k,0}. \quad \text{(S1.47)}$$

Case 6: $\mathbf{h}_{\mu\nu\kappa} = \mathbf{h}_{i-1jk-1}$

In the case that $\mathbf{h}_{\mu\nu\kappa} = \mathbf{h}_{i-1jk-1}$, $\boldsymbol{r}_{101}$ in Eq. (S1.4) corresponds with $\boldsymbol{r}_{ijk}$. As a result:

$$\frac{\partial \mathbf{h}_{i-1jk-1}}{\partial x_{ijk}} = \frac{1}{4}\begin{bmatrix} 1 & 0 & 0 \\ -1 & 0 & 0 \\ 1 & 0 & 0 \end{bmatrix} \qquad \frac{\partial \mathbf{h}_{i-1jk-1}}{\partial y_{ijk}} = \frac{1}{4}\begin{bmatrix} 0 & 1 & 0 \\ 0 & -1 & 0 \\ 0 & 1 & 0 \end{bmatrix} \qquad \frac{\partial \mathbf{h}_{i-1jk-1}}{\partial z_{ijk}} = \frac{1}{4}\begin{bmatrix} 0 & 0 & 1 \\ 0 & 0 & -1 \\ 0 & 0 & 1 \end{bmatrix}. \tag{S1.48}$$

As a consequence, we find for the strain derivative:

$$\begin{aligned}
\frac{\partial \varepsilon_{i-1jk-1}}{\partial x_{ijk}} &= \frac{1}{8}\mathbf{h}^{-T}_{i-1jk-1,0}\left(\begin{bmatrix} 1 & -1 & 1 \\ 0 & 0 & 0 \\ 0 & 0 & 0 \end{bmatrix}\mathbf{h}_{i-1jk-1} + \mathbf{h}^{T}_{i-1jk-1}\begin{bmatrix} 1 & 0 & 0 \\ -1 & 0 & 0 \\ 1 & 0 & 0 \end{bmatrix}\right)\mathbf{h}^{-1}_{i-1jk-1,0} && \text{(S1.49)} \\
&= \frac{1}{32}\mathbf{h}^{-T}_{i-1jk-1,0}\left(\begin{bmatrix} f_6(x_{ijk}) & f_6(y_{ijk}) & f_6(z_{ijk}) \\ 0 & 0 & 0 \\ 0 & 0 & 0 \end{bmatrix} + \begin{bmatrix} f_6(x_{ijk}) & 0 & 0 \\ f_6(y_{ijk}) & 0 & 0 \\ f_6(z_{ijk}) & 0 & 0 \end{bmatrix}\right)\mathbf{h}^{-1}_{i-1jk-1,0} && \text{(S1.50)} \\
&= \frac{1}{32}\mathbf{h}^{-T}_{i-1jk-1,0}\begin{bmatrix} 2f_6(x_{ijk}) & f_6(y_{ijk}) & f_6(z_{ijk}) \\ f_6(y_{ijk}) & 0 & 0 \\ f_6(z_{ijk}) & 0 & 0 \end{bmatrix}\mathbf{h}^{-1}_{i-1jk-1,0}. && \text{(S1.51)}
\end{aligned}$$

with

$$f_6(x_{ijk}) = -x_{i-1jk-1} + x_{ijk-1} - 3x_{i-1j+1k-1} + x_{i-1jk} - x_{ij+1k-1} + 3x_{ijk} - x_{i-1j+1k} + x_{ij+1k}. \tag{S1.52}$$

Similarly

$$\frac{\partial \varepsilon_{i-1jk-1}}{\partial y_{ijk}} = \frac{1}{32}\mathbf{h}^{-T}_{i-1jk-1,0}\begin{bmatrix} 0 & f_6(x_{ijk}) & 0 \\ f_6(x_{ijk}) & 2f_6(y_{ijk}) & f_6(z_{ijk}) \\ 0 & f_6(z_{ijk}) & 0 \end{bmatrix}\mathbf{h}^{-1}_{i-1jk-1,0} \tag{S1.53}$$

and

$$\frac{\partial \varepsilon_{i-1jk-1}}{\partial z_{ijk}} = \frac{1}{32}\mathbf{h}_{i-1jk-1,0}^{-T}\begin{bmatrix} 0 & 0 & f_6(x_{ijk}) \\ 0 & 0 & f_6(y_{ijk}) \\ f_6(x_{ijk}) & f_6(y_{ijk}) & 2f_6(z_{ijk}) \end{bmatrix}\mathbf{h}_{i-1jk-1,0}^{-1}. \tag{S1.54}$$

Case 7: $\mathbf{h}_{\mu\nu\kappa} = \mathbf{h}_{ij-1k-1}$

In the case that $\mathbf{h}_{\mu\nu\kappa} = \mathbf{h}_{ij-1k-1}$, $\boldsymbol{r}_{011}$ in Eq. (S1.4) corresponds with $\boldsymbol{r}_{ijk}$. As a result:

$$\frac{\partial \mathbf{h}_{ij-1k-1}}{\partial x_{ijk}} = \frac{1}{4}\begin{bmatrix} -1 & 0 & 0 \\ 1 & 0 & 0 \\ 1 & 0 & 0 \end{bmatrix} \quad \frac{\partial \mathbf{h}_{ij-1k-1}}{\partial y_{ijk}} = \frac{1}{4}\begin{bmatrix} 0 & -1 & 0 \\ 0 & 1 & 0 \\ 0 & 1 & 0 \end{bmatrix} \quad \frac{\partial \mathbf{h}_{ij-1k-1}}{\partial z_{ijk}} = \frac{1}{4}\begin{bmatrix} 0 & 0 & -1 \\ 0 & 0 & 1 \\ 0 & 0 & 1 \end{bmatrix}. \tag{S1.55}$$

As a consequence, we find for the strain derivative:

$$\begin{aligned} \frac{\partial \varepsilon_{ij-1k-1}}{\partial x_{ijk}} &= \frac{1}{8}\mathbf{h}_{ij-1k-1,0}^{-T}\left(\begin{bmatrix} -1 & 1 & 1 \\ 0 & 0 & 0 \\ 0 & 0 & 0 \end{bmatrix}\mathbf{h}_{ij-1k-1} + \mathbf{h}_{ij-1k-1}^{T}\begin{bmatrix} -1 & 0 & 0 \\ 1 & 0 & 0 \\ 1 & 0 & 0 \end{bmatrix}\right)\mathbf{h}_{ij-1k-1,0}^{-1} & \text{(S1.56)} \\ &= \frac{1}{32}\mathbf{h}_{ij-1k-1,0}^{-T}\left(\begin{bmatrix} f_7(x_{ijk}) & f_7(y_{ijk}) & f_7(z_{ijk}) \\ 0 & 0 & 0 \\ 0 & 0 & 0 \end{bmatrix} + \begin{bmatrix} f_7(x_{ijk}) & 0 & 0 \\ f_7(y_{ijk}) & 0 & 0 \\ f_7(z_{ijk}) & 0 & 0 \end{bmatrix}\right)\mathbf{h}_{ij-1k-1,0}^{-1} & \text{(S1.57)} \\ &= \frac{1}{32}\mathbf{h}_{ij-1k-1,0}^{-T}\begin{bmatrix} 2f_7(x_{ijk}) & f_7(y_{ijk}) & f_7(z_{ijk}) \\ f_7(y_{ijk}) & 0 & 0 \\ f_7(z_{ijk}) & 0 & 0 \end{bmatrix}\mathbf{h}_{ij-1k-1,0}^{-1}. & \text{(S1.58)} \end{aligned}$$

with

$$f_7(x_{ijk}) = -x_{ij-1k-1} - 3x_{i+1j-1k-1} + x_{ijk-1} + x_{ij-1k} - x_{i+1jk-1} - x_{i+1j-1k} + 3x_{ijk} + x_{i+1jk}. \tag{S1.59}$$

Similarly

$$\frac{\partial \varepsilon_{ij-1k-1}}{\partial y_{ijk}} = \frac{1}{32}\mathbf{h}_{ij-1k-1,0}^{-T} \begin{bmatrix} 0 & f_7(x_{ijk}) & 0 \\ f_7(x_{ijk}) & 2f_7(y_{ijk}) & f_7(z_{ijk}) \\ 0 & f_7(z_{ijk}) & 0 \end{bmatrix} \mathbf{h}_{ij-1k-1,0}^{-1} \tag{S1.60}$$

and

$$\frac{\partial \varepsilon_{ij-1k-1}}{\partial z_{ijk}} = \frac{1}{32}\mathbf{h}_{ij-1k-1,0}^{-T} \begin{bmatrix} 0 & 0 & f_7(x_{ijk}) \\ 0 & 0 & f_7(y_{ijk}) \\ f_7(x_{ijk}) & f_7(y_{ijk}) & 2f_7(z_{ijk}) \end{bmatrix} \mathbf{h}_{ij-1k-1,0}^{-1}. \tag{S1.61}$$

Case 8: $\mathbf{h}_{\mu\nu\kappa} = \mathbf{h}_{i-1j-1k-1}$

In the case that $\mathbf{h}_{\mu\nu\kappa} = \mathbf{h}_{i-1j-1k-1}$, $\boldsymbol{r}_{111}$ in Eq. (S1.4) corresponds with $\boldsymbol{r}_{ijk}$. As a result:

$$\frac{\partial \mathbf{h}_{i-1j-1k-1}}{\partial x_{ijk}} = \frac{1}{4}\begin{bmatrix} 1 & 0 & 0 \\ 1 & 0 & 0 \\ 1 & 0 & 0 \end{bmatrix} \quad \frac{\partial \mathbf{h}_{i-1j-1k-1}}{\partial y_{ijk}} = \frac{1}{4}\begin{bmatrix} 0 & 1 & 0 \\ 0 & 1 & 0 \\ 0 & 1 & 0 \end{bmatrix} \quad \frac{\partial \mathbf{h}_{i-1j-1k-1}}{\partial z_{ijk}} = \frac{1}{4}\begin{bmatrix} 0 & 0 & 1 \\ 0 & 0 & 1 \\ 0 & 0 & 1 \end{bmatrix}. \tag{S1.62}$$

As a consequence, we find for the strain derivative:

$$\frac{\partial \varepsilon_{i-1j-1k-1}}{\partial x_{ijk}} = \frac{1}{8}\mathbf{h}^{-T}_{i-1j-1k-1,0}\left(\begin{bmatrix} 1 & 1 & 1 \\ 0 & 0 & 0 \\ 0 & 0 & 0 \end{bmatrix}\mathbf{h}_{i-1j-1k-1} + \mathbf{h}^{T}_{i-1j-1k-1}\begin{bmatrix} 1 & 0 & 0 \\ 1 & 0 & 0 \\ 1 & 0 & 0 \end{bmatrix}\right)\mathbf{h}^{-1}_{i-1j-1k-1,0} \tag{S1.63}$$

$$= \frac{1}{32}\mathbf{h}^{-T}_{i-1j-1k-1,0}\left(\begin{bmatrix} f_8(x_{ijk}) & f_8(y_{ijk}) & f_8(z_{ijk}) \\ 0 & 0 & 0 \\ 0 & 0 & 0 \end{bmatrix} + \begin{bmatrix} f_8(x_{ijk}) & 0 & 0 \\ f_8(y_{ijk}) & 0 & 0 \\ f_8(z_{ijk}) & 0 & 0 \end{bmatrix}\right)\mathbf{h}^{-1}_{i-1j-1k-1,0} \tag{S1.64}$$

$$= \frac{1}{32}\mathbf{h}^{-T}_{i-1j-1k-1,0}\begin{bmatrix} 2f_8(x_{ijk}) & f_8(y_{ijk}) & f_8(z_{ijk}) \\ f_8(y_{ijk}) & 0 & 0 \\ f_8(z_{ijk}) & 0 & 0 \end{bmatrix}\mathbf{h}^{-1}_{i-1j-1k-1,0}. \tag{S1.65}$$

with

$$f_8(x_{ijk}) = -3x_{i-1j-1k-1} - x_{ij-1k-1} - x_{i-1jk-1} - x_{i-1j-1k} + x_{ijk-1} + x_{ij-1k} + x_{i-1jk} + 3x_{ijk}. \tag{S1.66}$$

Similarly

$$\frac{\partial \varepsilon_{i-1j-1k-1}}{\partial y_{ijk}} = \frac{1}{32}\mathbf{h}^{-T}_{i-1j-1k-1,0}\begin{bmatrix} 0 & f_8(x_{ijk}) & 0 \\ f_8(x_{ijk}) & 2f_8(y_{ijk}) & f_8(z_{ijk}) \\ 0 & f_8(z_{ijk}) & 0 \end{bmatrix}\mathbf{h}^{-1}_{i-1j-1k-1,0} \tag{S1.67}$$

and

$$\frac{\partial \varepsilon_{i-1j-1k-1}}{\partial z_{ijk}} = \frac{1}{32}\mathbf{h}^{-T}_{i-1j-1k-1,0}\begin{bmatrix} 0 & 0 & f_8(x_{ijk}) \\ 0 & 0 & f_8(y_{ijk}) \\ f_8(x_{ijk}) & f_8(y_{ijk}) & 2f_8(z_{ijk}) \end{bmatrix}\mathbf{h}^{-1}_{i-1j-1k-1,0}. \tag{S1.68}$$

S1.4 Resulting equations of motion for unistable periodic systems

In conclusion, the update equations for the unistable periodic micromechanical system are given by $\dot{\boldsymbol{r}}_{ijk} = \boldsymbol{p}_{ijk}/m_{ijk} = \boldsymbol{v}_{ijk}$ and

$$
\begin{aligned}
\dot{p}_{ijk,x} = & -\frac{1}{2}\sum_{\mu=i-1}^{i}\sum_{\nu=j-1}^{j}\sum_{\kappa=k-1}^{k}\det(\mathbf{h}_{\mu\nu\kappa})\left[\mathrm{Tr}\left\{\mathbf{h}_{\mu\nu\kappa}^{-1}\frac{\partial}{\partial x_{ijk}}\mathbf{h}_{\mu\nu\kappa}\right\}\boldsymbol{\varepsilon}_{\mu\nu\kappa}^{T}:\mathbf{C}_{\mu\nu\kappa,0}:\boldsymbol{\varepsilon}_{\mu\nu\kappa}\right.\\
& +\frac{1}{32}\left(\mathbf{h}_{\mu\nu\kappa,0}^{-T}\begin{bmatrix} 2f_{n_{\mu\nu\kappa}}(x_{ijk}) & f_{n_{\mu\nu\kappa}}(y_{ijk}) & f_{n_{\mu\nu\kappa}}(a_{ijk}) \\ f_{n_{\mu\nu\kappa}}(y_{ijk}) & 0 & 0 \\ f_{n_{\mu\nu\kappa}}(z_{ijk}) & 0 & 0 \end{bmatrix}\mathbf{h}_{\mu\nu\kappa,0}^{-1}\right):\mathbf{C}_{\mu\nu\kappa,0}:\boldsymbol{\varepsilon}_{\mu\nu\kappa}\\
& \left.+\frac{1}{32}\,\boldsymbol{\varepsilon}_{\mu\nu\kappa}^{T}:\mathbf{C}_{\mu\nu\kappa,0}:\left(\mathbf{h}_{\mu\nu\kappa,0}^{-T}\begin{bmatrix} 2f_{n_{\mu\nu\kappa}}(x_{ijk}) & f_{n_{\mu\nu\kappa}}(y_{ijk}) & f_{n_{\mu\nu\kappa}}(z_{ijk}) \\ f_{n_{\mu\nu\kappa}}(y_{ijk}) & 0 & 0 \\ f_{n_{\mu\nu\kappa}}(z_{ijk}) & 0 & 0 \end{bmatrix}\mathbf{h}_{\mu\nu\kappa,0}^{-1}\right)\right] \qquad \text{(S1.69)}
\end{aligned}
$$

$$
\begin{aligned}
\dot{p}_{ijk,y} = & -\frac{1}{2}\sum_{\mu=i-1}^{i}\sum_{\nu=j-1}^{j}\sum_{\kappa=k-1}^{k}\det(\mathbf{h}_{\mu\nu\kappa})\left[\mathrm{Tr}\left\{\mathbf{h}_{\mu\nu\kappa}^{-1}\frac{\partial}{\partial y_{ijk}}\mathbf{h}_{\mu\nu\kappa}\right\}\boldsymbol{\varepsilon}_{\mu\nu\kappa}^{T}:\mathbf{C}_{\mu\nu\kappa,0}:\boldsymbol{\varepsilon}_{\mu\nu\kappa}\right.\\
& +\frac{1}{32}\left(\mathbf{h}_{\mu\nu\kappa,0}^{-T}\begin{bmatrix} 0 & f_{n_{\mu\nu\kappa}}(x_{ijk}) & 0 \\ f_{n_{\mu\nu\kappa}}(x_{ijk}) & 2f_{n_{\mu\nu\kappa}}(y_{ijk}) & f_{n_{\mu\nu\kappa}}(z_{ijk}) \\ 0 & f_{n_{\mu\nu\kappa}}(z_{ijk}) & 0 \end{bmatrix}\mathbf{h}_{\mu\nu\kappa,0}^{-1}\right):\mathbf{C}_{\mu\nu\kappa,0}:\boldsymbol{\varepsilon}_{\mu\nu\kappa}\\
& \left.+\frac{1}{32}\,\boldsymbol{\varepsilon}_{\mu\nu\kappa}^{T}:\mathbf{C}_{\mu\nu\kappa,0}:\left(\mathbf{h}_{\mu\nu\kappa,0}^{-T}\begin{bmatrix} 0 & f_{n_{\mu\nu\kappa}}(x_{ijk}) & 0 \\ f_{n_{\mu\nu\kappa}}(x_{ijk}) & 2f_{n_{\mu\nu\kappa}}(y_{ijk}) & f_{n_{\mu\nu\kappa}}(z_{ijk}) \\ 0 & f_{n_{\mu\nu\kappa}}(z_{ijk}) & 0 \end{bmatrix}\mathbf{h}_{\mu\nu\kappa,0}^{-1}\right)\right] \qquad \text{(S1.70)}
\end{aligned}
$$

$$
\begin{aligned}
\dot{p}_{ijk,z} \quad = \quad & -\frac{1}{2}\sum_{\mu=i-1}^{i}\sum_{\nu=j-1}^{j}\sum_{\kappa=k-1}^{k}\det(\mathbf{h}_{\mu\nu\kappa})\left[\mathrm{Tr}\left\{\mathbf{h}_{\mu\nu\kappa}^{-1}\frac{\partial}{\partial z_{ijk}}\mathbf{h}_{\mu\nu\kappa}\right\}\boldsymbol{\varepsilon}_{\mu\nu\kappa}^{T}:\mathbf{C}_{\mu\nu\kappa,0}:\boldsymbol{\varepsilon}_{\mu\nu\kappa}\right. \\
& +\frac{1}{32}\left(\mathbf{h}_{\mu\nu\kappa,0}^{-T}\begin{bmatrix} 0 & 0 & f_{n_{\mu\nu\kappa}}(x_{ijk}) \\ 0 & 0 & f_{n_{\mu\nu\kappa}}(y_{ijk}) \\ f_{n_{\mu\nu\kappa}}(x_{ijk}) & f_{n_{\mu\nu\kappa}}(y_{ijk}) & 2f_{n_{\mu\nu\kappa}}(z_{ijk}) \end{bmatrix}\mathbf{h}_{\mu\nu\kappa,0}^{-1}\right):\mathbf{C}_{\mu\nu\kappa,0}:\boldsymbol{\varepsilon}_{\mu\nu\kappa} \\
& \left.+\frac{1}{32}\,\boldsymbol{\varepsilon}_{\mu\nu\kappa}^{T}:\mathbf{C}_{\mu\nu\kappa,0}:\left(\mathbf{h}_{\mu\nu\kappa,0}^{-T}\begin{bmatrix} 0 & 0 & f_{n_{\mu\nu\kappa}}(x_{ijk}) \\ 0 & 0 & f_{n_{\mu\nu\kappa}}(y_{ijk}) \\ f_{n_{\mu\nu\kappa}}(x_{ijk}) & f_{n_{\mu\nu\kappa}}(y_{ijk}) & 2f_{n_{\mu\nu\kappa}}(z_{ijk}) \end{bmatrix}\mathbf{h}_{\mu\nu\kappa,0}^{-1}\right)\right] \qquad \text{(S1.71)}
\end{aligned}
$$

The first-order polynomials $f_{n_{\mu\nu\kappa}}$ as well as the derivatives of $\mathbf{h}_{\mu\nu\kappa}$ appearing in these update equations were determined in Section S1.3 for each of the eight possible (μ, ν, κ) values entering the summation of Eq. (S1.9).

S1.5 Extension to unistable isolated systems

While the equations of motion derived in Section S1.4 are valid for a periodic 3D system, they can be easily extended to also describe an isolated micromechanical system. To this end, consider again an $n_x \times n_y \times n_z$ collection of nanocells, each defined by the eight nodes on their corners. For a periodic system, we identified for instance $\boldsymbol{r}_{n_x jk}$ with $\boldsymbol{r}_{0jk}$. For an isolated system, however, all nodes on the boundary, *i.e.*, nodes N_{ijk} with $i \in \{0, n_x\}$, $j \in \{0, n_y\}$ or $k \in \{0, n_z\}$ are unique. Therefore, $(n_x + 1) \times (n_y + 1) \times (n_z + 1)$ nodes are needed to define the system, resulting in the following Lagrangian and Hamiltonian:

$$
\mathcal{L}(\boldsymbol{r}, \dot{\boldsymbol{r}}) = \sum_{i=0}^{n_x}\sum_{j=0}^{n_y}\sum_{k=0}^{n_z}\frac{1}{2}m_{ijk}\dot{\boldsymbol{r}}_{ijk}^2 - \sum_{\mu=0}^{n_x-1}\sum_{\nu=0}^{n_y-1}\sum_{\kappa=0}^{n_z-1}\mathcal{U}_{\mu\nu\kappa}(\boldsymbol{r}); \qquad \text{(S1.72)}
$$

$$
\mathcal{H}(\boldsymbol{r}, \boldsymbol{p}) = \sum_{i=0}^{n_x}\sum_{j=0}^{n_y}\sum_{k=0}^{n_z}\frac{\boldsymbol{p}_{ijk}^2}{2m_{ijk}} + \sum_{\mu=0}^{n_x-1}\sum_{\nu=0}^{n_y-1}\sum_{\kappa=0}^{n_z-1}\mathcal{U}_{\mu\nu\kappa}(\boldsymbol{r}). \qquad \text{(S1.73)}
$$

The interpretation of all these variables is the same as in the periodic system. Especially,

m_{ijk} still denotes the mass assigned to node N_{ijk}, which is typically taken as the average of the eight nanocells surrounding the node. In the case that the node N_{ijk} is found on the boundary of the system, a zero mass is assigned to each of the in total eight terms entering this averaging procedure that fall outside the isolated model. This ensures that the nodes on the boundary generally have a lower mass and a larger flexibility compared to bulk nodes, as can be intuitively expected for an isolated system.

Given that the functional form of the Hamiltonian remains the same, also the update equations of the isolated system remain unaltered with respect to those determined for the periodic system in Section S1.4; keeping in mind that the update equations for the momentum $\boldsymbol{p}_{ijk}$ will contain fewer than eight terms in the case that a boundary node is considered as no elastic energy is associated with the vacuum surrounding the isolated system.

S2 Extension towards bistable nanocells

As outlined in the main text, the micromechanical model derived for unistable nanocells can be easily extended to account for bistable nanocells by using the interpolation scheme of Christ and van Gunsteren.[1] This interpolation scheme is introduced in Section S2.1 and result in the equations of motion of Section S2.2.

S2.1 Interpolation of the elastic deformation energy

Using the interpolation scheme of Ref. 1, the potential energy for a bistable nanocell $\mathbf{h}_{\mu\nu\kappa}$ is given by

$$\mathcal{U}_{\mu\nu\kappa} = -k_B T^*_{\mu\nu\kappa} \ln \left[\exp\left(-\frac{\mathcal{U}^{(I)}_{\mu\nu\kappa}}{k_B T^*_{\mu\nu\kappa}} \right) + \exp\left(-\frac{\mathcal{U}^{(II)}_{\mu\nu\kappa} + \Delta F_{I\leftrightarrow II,\mu\nu\kappa}}{k_B T^*_{\mu\nu\kappa}} \right) \right]. \tag{S2.1}$$

In this expression, $\mathcal{U}^{(I)}_{\mu\nu\kappa}$ and $\mathcal{U}^{(II)}_{\mu\nu\kappa}$ are the single-phase elastic deformation energies, determined based on the equilibrium cell matrix and stiffness tensor of these phases as outlined in Eq. (S1.2), and $\Delta F_{I\leftrightarrow II,\mu\nu\kappa}$ is the difference in free energy between the two phases, determined from the atomistic simulation. Furthermore, k_B is the Boltzmann constant and $T^*_{\mu\nu\kappa}$ is an effective temperature that determines the smoothness of the interpolation. In practice, this parameter should be fitted to best reproduce the barrier $\Delta F_{b,\mu\nu\kappa}$ obtained from atomistic simulations (see Figure 4 in the main text for a visual representation).

S2.2 Resulting equations of motion for bistable systems

From the equations of motion for the unistable system, given in Eq. (S1.8) and Eq. (S1.9), it is clear that the altered potential energy will only directly affect the update equation for the conjugated momenta, through $\nabla_{\boldsymbol{r}_{ijk}} \mathcal{U}_{\mu\nu\kappa}(\boldsymbol{r})$. From Eq. (S2.1), we find for the bistable nanocell that

$$\nabla_{\boldsymbol{r}_{ijk}} \mathcal{U}_{\mu\nu\kappa}(\boldsymbol{r}) = \frac{\exp\left(-\frac{\mathcal{U}^{(I)}_{\mu\nu\kappa}(\boldsymbol{r})}{k_B T^*_{\mu\nu\kappa}} \right) \nabla_{\boldsymbol{r}_{ijk}} \mathcal{U}^{(I)}_{\mu\nu\kappa}(\boldsymbol{r}) + \exp\left(-\frac{\mathcal{U}^{(II)}_{\mu\nu\kappa}(\boldsymbol{r}) + \Delta F_{I\leftrightarrow II,\mu\nu\kappa}}{k_B T^*_{\mu\nu\kappa}} \right) \nabla_{\boldsymbol{r}_{ijk}} \mathcal{U}^{(II)}_{\mu\nu\kappa}(\boldsymbol{r})}{\exp\left(-\frac{\mathcal{U}^{(I)}_{\mu\nu\kappa}(\boldsymbol{r})}{k_B T^*_{\mu\nu\kappa}} \right) + \exp\left(-\frac{\mathcal{U}^{(II)}_{\mu\nu\kappa}(\boldsymbol{r}) + \Delta F_{I\leftrightarrow II,\mu\nu\kappa}}{k_B T^*_{\mu\nu\kappa}} \right)}. \tag{S2.2}$$

The derivatives $\nabla_{\boldsymbol{r}_{ijk}} \mathcal{U}^{(I)}_{\mu\nu\kappa}(\boldsymbol{r})$ and $\nabla_{\boldsymbol{r}_{ijk}} \mathcal{U}^{(II)}_{\mu\nu\kappa}(\boldsymbol{r})$ that enter this equation are derivatives of the single-phase elastic deformation energy. As they were explicitly derived in Section S1.3, they can be directly plugged in in Eq. (S2.2) to yield the update equation for the conjugated momenta of nodes that define bistable nanocells.

In the update equation of the conjugated momentum of node N_{ijk},

$$\dot{\boldsymbol{p}}_{ijk} = -\sum_{\mu=i-1}^{i}\sum_{\nu=j-1}^{j}\sum_{\kappa=k-1}^{k}\nabla_{\boldsymbol{r}_{ijk}}\mathcal{U}_{\mu\nu\kappa}(\boldsymbol{r}), \tag{S2.3}$$

each of the eight terms $\nabla_{\boldsymbol{r}_{ijk}}\mathcal{U}_{\mu\nu\kappa}(\boldsymbol{r})$ will thus correspond with the derivatives calculated in Section S1.3 if that specific nanocell is unistable, or correspond with Eq. (S2.2) if that specific nanocell is bistable. To extend these update equations to isolated systems, the same procedure as in Section S1.5 for unistable systems can be followed.

References

(1) Christ, C. D.; van Gunsteren, W. F. Enveloping distribution sampling: A method to calculate free energy differences from a single simulation. *J. Chem. Phys.* **2007**, *126*, 184110.