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Atomistic two-, three- and four-body potentials. Spatial and material settings

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ABSTRACT

In molecular dynamics or molecular statics (MD/MS) multi-body potentials empirically capture the energetic interactions in atomistic systems enabling the computation of the corresponding atomistic forces as energetic conjugates to the atomistic positions. We distinguish here between spatial and material atomistic positions and consequently between the corresponding spatial and material atomistic forces. In quasi-statics, i.e. MS, the former, also denoted as *deformational* atomistic forces, contribute to the classical deformational mechanics (i.e., equilibrium) problem that seeks to minimise the total potential energy of an atomistic system with respect to the atomistic positions relative to the ambient space. The latter, also denoted as *configurational* atomistic forces, contribute to the configurational mechanics (i.e., non-equilibrium) problem that determines the release of total potential energy of an atomistic system upon variation of the atomistic positions relative to the ambient material, i.e., due to perturbations of the material (initial) atomistic configuration. The importance of material atomistic forces is that they drive energetically favourable re-organisations of the material atomistic configuration, thereby characterising the tendency of generic atomistic defects to propagate. In this contribution we focus on two-, three-, and four-body potentials, whereby we distinguish between novel stretch- and classical angle-based potentials for the two latter cases. Taken together, as the main contribution, we derive expressions for the corresponding spatial and, for the first time, material atomistic forces and highlight their striking formal similarity. The derivations are detailed but the final expression compact and well-suited for numerical implementation.

1. Introduction

When modelling the energetic interactions in atomistic systems by molecular dynamics or molecular statics (MD/MS), multi-body potentials are a valid option to empirically approximate the complex energy landscape that is dictated by the underlying quantum mechanics. Most prominent of the numerous options for two-body potentials are the Buckingham and Lennard-Jones (1938) and the Lennard-Jones (1924a,b, 1925) potentials, which only account for the distance between atomistic *pairs*. Well-known among higher-order models is the Stillinger–Weber potential (Stillinger and Weber, 1985) which combines two- and three-body terms. A further typical example of a three-body potential is due to Tersoff (1988), which is a prominent member of the class of bond-order potentials that are motivated by the tight-binding approximation, see Albe et al. (2002). The Tersoff potential parameterises the atomistic energy by the distance between atomistic pairs and, in addition, includes atomistic *triplets* through the coefficients of a two-body term. Another common choice of a multi-body potential is the Embedded Atom potential (Daw and Baskes, 1984), which besides the distance between atomistic pairs accounts also for the embedding electron charge density of neighbouring atoms. Many approaches that rely on simple harmonic potentials to describe atomistic interactions are complemented by three-body terms parameterising the energetics of fluctuations around an average angle between atomistic triplets, typically based on a simple harmonic approximation. Furthermore, force fields, see e.g., Brooks et al. (1983) and Cornell et al. (1995) are complemented by four-body terms incorporating

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dihedral angle fluctuations, i.e. the torsional rotation of atomistic *tetrads* about a central bond. However, while the corresponding parametrisation typically involves the cosine map of the dihedral angle, the specific format of the potentials is very much model dependent, see for example the incomplete list of models (Hess et al., 2008; Plimpton, 1995; Mayo et al., 1990; Allinger et al., 1989, 2003; Bureekaew et al., 2013; Rappe et al., 1992; Jorgensen and Tirado-Rives, 2005; Wang et al., 2004; Yildirim et al., 2010).

Conceptually, two-, three- and four-body potentials capture the energetic interactions between any potentially possible combination of corresponding atomistic pairs, triplets and tetrads within an atomistic system. In practise, the plethora of potentially possible combinations is restricted by interaction cut-offs. It is important to note that we shall not consider cut-offs here for the sake of presentation.

Whereas objectivity (invariance under superposed rigid body motions) and parity symmetry (invariance under inversion of space) restricts the parametrisation of two-body (pair) potentials to the (scalar-valued) length of an atomistic pair, see Tadmor and Miller (2011), three- and four-body (triplet and tetrad, respectively) potentials allow for two different types of parametrisation. Motivated in part by the intuitive concept of rotational springs, three- and four-body potentials are commonly parameterised in terms of angles between adjacent atomistic pairs and dihedral angles between adjacent atomistic triplets. However, inspired by continuum kinematics that involves the maps of line, area and volume elements as local measures of deformation, an alternative parametrisation in terms of triplet areas and tetrad volumes is another valid option.

Similarly inspired by continuum kinematics and following our earlier work on pair potentials (Steinmann et al., 2011; Birang O. and Steinmann, 2021), we introduce pair, triplet and tetrad *stretches* rather than pair lengths, triplet areas and tetrad volumes. Thereby, we use the terminology pair, triplet and tetrad stretch to denote the ratio of either the pair lengths, triplet areas or tetrad volumes in the deformed and undeformed state, respectively, of an atomistic system. In analogy to the continuum setting, we denote these states of an atomistic system as the *spatial* and *material* atomistic configurations, respectively. In the same vein, we introduce the ratio of the triplet and tetrad angles in the spatial and material atomistic configurations as the triplet and tetrad *twist*, respectively, whereby we use, for convenience, the re-parametrisation of angles in terms of the cosine map (avoiding singularities for almost all practical cases, i.e. only excluding extremely unlikely right angles).

Parameterising two-, three- and four-body potentials either in terms of pair, triplet and tetrad stretches or in terms of pair stretch as well as triplet and tetrad twists proves particularly beneficial when elaborating and contrasting the corresponding spatial and material atomistic forces deriving from the various options for multi-body potentials. In the quasi-static MS setting considered here, spatial atomistic forces contribute to the classical *deformational* (equilibrium) problem that seeks to minimise the total potential energy of an atomistic system when varying the spatial atomistic positions. By contrast, material atomistic forces contribute to the *configurational* (non-equilibrium) problem that determines the release of total potential energy of an atomistic system when varying the material (initial) atomistic positions. Thereby, material atomistic forces characterise the tendency of generic atomistic defects to propagate, i.e., they drive energetically favourable re-organisations of the material atomistic configuration.

As our main contribution, we demonstrate, based on a systematic and unifying approach to the kinematics (and energetics) of atomistic pairs, triplets and tetrads, the striking formal similarity of the resulting spatial and material atomistic forces. Notably, this format also compares well to the energy–momentum structure of the so-called Eshelby stress in the continuum setting of configurational mechanics, see Eshelby (1975). Views on the continuum setting of configurational mechanics are expressed for example in Maugin (1993, 1995, 2011), Gurtin (1995, 2000), Cermelli and Fried (1997) and Fried and Gurtin (2003). Our own contributions to configurational mechanics are documented, for example, in Steinmann (2000, 2002b,a,c, 2008), Steinmann et al. (2009) and Steinmann et al. (2011).

Moreover, attempts towards the transition from discrete to continuous descriptions of matter, as for example Findeisen et al. (2020), Turco et al. (2016), Davydov and Steinmann (2014a,b, 2015) (among others), will clearly benefit from our systematic and unifying approach when taking into account multi-body interactions at the discrete level.

Finally, the advocated multi-body formulation for the MS of atomistic systems not only highlights the fundamental duality between a stretch-based and a twist-based approach, but it also reveals an underlying conceptual similarity between the stretch-based atomistic formulation and the *peridynamics* formulation recently proposed by the authors Javili et al. (2019, 2020, 2021c,a,b). Furthermore, the current manuscript suggests that a parallel twist-based framework for peridynamics can also be established.

The remainder of the manuscript is organised as follows. Section 2 comprehensively details the kinematics of atomistic systems pertinent to atomistic pairs, triplets and tetrads, carefully distinguishing between the stretch-based and twist-based approach in the two latter cases. Section 3 addresses first the equilibrium of spatial atomistic forces within MS as a consequence of total energy minimisation within the atomistic setting of deformational mechanics. The focus thereafter is on the non-equilibrium of material atomistic forces driving energy release within the atomistic setting of configurational mechanics. Section 4 derives concise and explicit representations for the spatial atomistic forces and contrasts their format for the stretch-based and twist-based parameterisations of the internal potential energy. In passing, it also touches on special cases with atomistic positions constrained to two- and one-dimensional manifolds (as relevant for 2d and 1d materials). Furthermore, it elaborates on the corresponding material atomistic forces and compares their format to that of the spatial atomistic forces. Section 5 closes the manuscript with conclusions and an outlook. The appendix assembles necessary, however lengthy and tedious, intermediate manipulations of atomistic expressions.

For the notation, we use blackboard font to symbolically denote vector- and tensor-valued atomistic quantities, meagre font to denote scalar-valued atomistic quantities and bold font to denote other vector- and tensor-valued quantities. Greek indices indicate the atomistic numbering, whereby we do not apply the summation convention on repeated Greek indices throughout the manuscript. *Full skew-symmetrisation* and *symmetrisation* (as detailed in the main text) of Greek index pairs, triplets and tetrads are denoted as $[\alpha\beta]$, $[\alpha\beta\gamma]$, $[\alpha\beta\gamma\delta]$ and $(\alpha\beta)$, $(\alpha\beta\gamma)$, $(\alpha\beta\gamma\delta)$, respectively. The sum of even permutations of Greek index triplets is denoted as $\langle\alpha\beta\gamma\rangle$.

Table 1
Summary of atomistic kinematics.

Atomistic pairs	Atomistic triplets	Atomistic tetrads
Pair length vector	Triplet area vector	Tetrad signed volume
$\mathbb{x}_{\alpha\beta} := \mathbb{x}_\beta - \mathbb{x}_\alpha$ $\mathbb{X}_{\alpha\beta} := \mathbb{X}_\beta - \mathbb{X}_\alpha$	$\mathbb{x}_{\alpha\beta\gamma} := \mathbb{x}_{\alpha\beta} \times \mathbb{x}_{\alpha\gamma}$ $\mathbb{X}_{\alpha\beta\gamma} := \mathbb{X}_{\alpha\beta} \times \mathbb{X}_{\alpha\gamma}$	$\mathsf{x}_{\alpha\beta\gamma\delta} := [\mathbb{x}_{\alpha\beta} \times \mathbb{x}_{\alpha\gamma}] \cdot \mathbb{x}_{\alpha\delta}$ $\mathsf{X}_{\alpha\beta\gamma\delta} := [\mathbb{X}_{\alpha\beta} \times \mathbb{X}_{\alpha\gamma}] \cdot \mathbb{X}_{\alpha\delta}$
Pair length	Triplet area	Tetrad volume
$x_{\alpha\beta} := \lvert\mathbb{x}_{\alpha\beta}\rvert$ $X_{\alpha\beta} := \lvert\mathbb{X}_{\alpha\beta}\rvert$	$x_{\alpha\beta\gamma} := \lvert\mathbb{x}_{\alpha\beta\gamma}\rvert$ $X_{\alpha\beta\gamma} := \lvert\mathbb{X}_{\alpha\beta\gamma}\rvert$	$x_{\alpha\beta\gamma\delta} := \lvert\mathsf{x}_{\alpha\beta\gamma\delta}\rvert$ $X_{\alpha\beta\gamma\delta} := \lvert\mathsf{X}_{\alpha\beta\gamma\delta}\rvert$
Pair direction	Triplet direction	Tetrad direction
$\mathbb{n}_{\alpha\beta} := \mathbb{x}_{\alpha\beta}/x_{\alpha\beta}$ $\mathbb{N}_{\alpha\beta} := \mathbb{X}_{\alpha\beta}/X_{\alpha\beta}$	$\mathbb{n}_{\alpha\beta\gamma} := \mathbb{x}_{\alpha\beta\gamma}/x_{\alpha\beta\gamma}$ $\mathbb{N}_{\alpha\beta\gamma} := \mathbb{X}_{\alpha\beta\gamma}/X_{\alpha\beta\gamma}$	$\mathsf{n}_{\alpha\beta\gamma\delta} := \mathsf{x}_{\alpha\beta\gamma\delta}/x_{\alpha\beta\gamma\delta}$ $\mathsf{N}_{\alpha\beta\gamma\delta} := \mathsf{X}_{\alpha\beta\gamma\delta}/X_{\alpha\beta\gamma\delta}$
Spatial atomistic sensitivities		
Pair-wise sensitivities	Triplet-wise sensitivities	Tetrad-wise sensitivities
$\partial x_{\alpha\beta}/\partial\mathbb{x}_{\alpha\beta} = \mathbb{n}_{\alpha\beta}$ $\partial\mathbb{n}_{\alpha\beta}/\partial\mathbb{x}_{\alpha\beta} = \mathbb{p}^\perp_{\alpha\beta}$	$\partial x_{\alpha\beta\gamma}/\partial\mathbb{x}_{\alpha\beta\gamma} = \mathbb{n}_{\alpha\beta\gamma}$ $\partial\mathbb{n}_{\alpha\beta\gamma}/\partial\mathbb{x}_{\alpha\beta\gamma} = \mathbb{p}^\perp_{\alpha\beta\gamma}$	$\partial x_{\alpha\beta\gamma\delta}/\partial\mathsf{x}_{\alpha\beta\gamma\delta} = \mathsf{n}_{\alpha\beta\gamma\delta}$ $\partial\mathsf{n}_{\alpha\beta\gamma\delta}/\partial\mathsf{x}_{\alpha\beta\gamma\delta} = \mathsf{p}^\perp_{\alpha\beta\gamma\delta}$
Fully symmetric orthogonal projection tensors		
$\mathbb{p}^\perp_{\alpha\beta} := [\mathbb{i} - \mathbb{n}_{\alpha\beta} \otimes \mathbb{n}_{\alpha\beta}]/x_{\alpha\beta}$	$\mathbb{p}^\perp_{\alpha\beta\gamma} := [\mathbb{i} - \mathbb{n}_{\alpha\beta\gamma} \otimes \mathbb{n}_{\alpha\beta\gamma}]/x_{\alpha\beta\gamma}$	$\mathsf{p}^\perp_{\alpha\beta\gamma\delta} := [\mathbb{1} - \mathsf{n}_{\alpha\beta\gamma\delta}\,\mathsf{n}_{\alpha\beta\gamma\delta}]/x_{\alpha\beta\gamma\delta}$
Pair stretch	Triplet stretch	Tetrad stretch
$\lambda_{\alpha\beta} := x_{\alpha\beta}/X_{\alpha\beta}$ $\Lambda_{\alpha\beta} := X_{\alpha\beta}/x_{\alpha\beta}$	$\lambda_{\alpha\beta\gamma} := x_{\alpha\beta\gamma}/X_{\alpha\beta\gamma}$ $\Lambda_{\alpha\beta\gamma} := X_{\alpha\beta\gamma}/x_{\alpha\beta\gamma}$	$\lambda_{\alpha\beta\gamma\delta} := x_{\alpha\beta\gamma\delta}/X_{\alpha\beta\gamma\delta}$ $\Lambda_{\alpha\beta\gamma\delta} := X_{\alpha\beta\gamma\delta}/x_{\alpha\beta\gamma\delta}$
	Triplet angle	Tetrad dihedral angle
	$\phi_{\alpha\beta\gamma} := \mathbb{n}_{\alpha\beta} \cdot \mathbb{n}_{\alpha\gamma}$ $\Phi_{\alpha\beta\gamma} := \mathbb{N}_{\alpha\beta} \cdot \mathbb{N}_{\alpha\gamma}$	$\phi_{\alpha\beta\gamma\delta} := \mathbb{n}_{\alpha\beta\gamma} \cdot \mathbb{n}_{\alpha\beta\delta}$ $\Phi_{\alpha\beta\gamma\delta} := \mathbb{N}_{\alpha\beta\gamma} \cdot \mathbb{N}_{\alpha\beta\delta}$
	Triplet twist	Tetrad dihedral twist
	$\omega_{\alpha\beta\gamma} := \phi_{\alpha\beta\gamma}/\Phi_{\alpha\beta\gamma}$ $\Omega_{\alpha\beta\gamma} := \Phi_{\alpha\beta\gamma}/\phi_{\alpha\beta\gamma}$	$\omega_{\alpha\beta\gamma\delta} := \phi_{\alpha\beta\gamma\delta}/\Phi_{\alpha\beta\gamma\delta}$ $\Omega_{\alpha\beta\gamma\delta} := \Phi_{\alpha\beta\gamma\delta}/\phi_{\alpha\beta\gamma\delta}$

2. Kinematics

The pertinent relations regarding the atomistic kinematics elaborated upon in this section are assembled for the sake of overview in Table 1.

2.1. Atomistic positions

An atomistic system consists of a large but finite number of atoms, labelled by α, that occupy the spatial and material *atomistic positions* $\mathbb{x}_\alpha$ and $\mathbb{X}_\alpha$, respectively, in a bounded domain of ambient space $\mathbb{E}^3$. These are related by a discrete, i.e., atom-wise motion with a time-like variable $s \in \mathbb{R}_+$, that here merely orders the quasi-static loading, see Fig. 1, as

$$\mathbb{x}_\alpha = \mathbb{x}_\alpha(s) \quad \text{with} \quad \mathbb{X}_\alpha := \mathbb{x}_\alpha(0). \tag{1}$$

In passing we remark that the spatial and material atomistic position-wise sensitivities render

$$\partial\mathbb{x}_\alpha/\partial\mathbb{x}_\alpha = \mathbb{i} \quad \text{and} \quad \partial\mathbb{X}_\alpha/\partial\mathbb{X}_\alpha = \mathbb{I},$$

with $\mathbb{i}$ and $\mathbb{I}$ denoting the second-order spatial and material identity tensors, respectively.

2.2. Atomistic pairs

2.2.1. Pair lengths

We introduce the spatial and material *pair length vectors* pointing from atom α to atom β, see Fig. 2, as

$$\mathbb{x}_{\alpha\beta} := \mathbb{x}_\beta - \mathbb{x}_\alpha \quad \text{and} \quad \mathbb{X}_{\alpha\beta} := \mathbb{X}_\beta - \mathbb{X}_\alpha. \tag{2}$$

Note the *skew-symmetry* in the indices of $\mathbb{x}_{\alpha\beta} = \mathbb{x}_{[\alpha\beta]}$ and $\mathbb{X}_{\alpha\beta} = \mathbb{X}_{[\alpha\beta]}$.

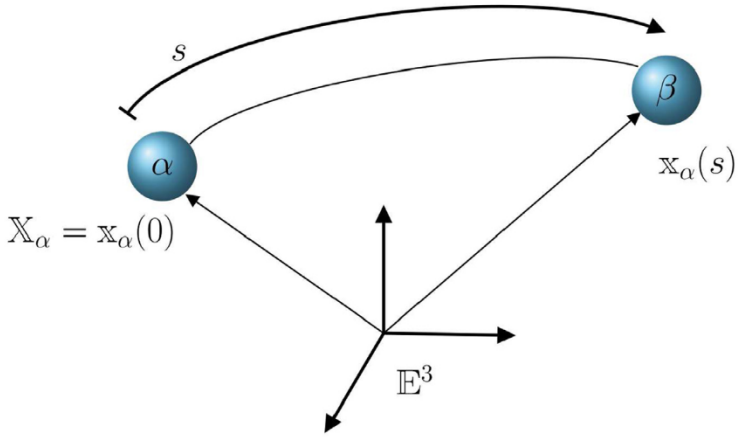


Fig. 1. Atomistic motion from material position at $s = 0$ to spatial position at s. The material atomistic positions are the positions the atoms take a time zero, i.e. before any external loading is applied. The spatial atomistic positions are then a result of the external loading.

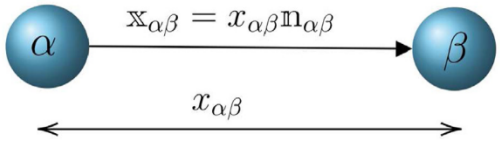


Fig. 2. Atomistic pair length vector and pair length.

In accordance with the requirement of *invariance*,[1] it is convenient to introduce the corresponding scalar-valued spatial and material *pair lengths* as

$$x_{\alpha\beta} := |\mathbb{x}_{\alpha\beta}| \quad \text{and} \quad X_{\alpha\beta} := |\mathbb{X}_{\alpha\beta}|. \tag{3}$$

Note the *symmetry* in the indices of $x_{\alpha\beta} = x_{(\alpha\beta)}$ and $X_{\alpha\beta} = X_{(\alpha\beta)}$, reflecting that the spatial and material pair lengths associated with each atom belonging to a specific atomistic pair are the same. Consequently, the vector-valued spatial and material *pair directions* (pair normals[2]) follow as

$$\mathbb{n}_{\alpha\beta} := \mathbb{x}_{\alpha\beta}/x_{\alpha\beta} \quad \text{and} \quad \mathbb{N}_{\alpha\beta} := \mathbb{X}_{\alpha\beta}/X_{\alpha\beta}. \tag{4}$$

Note the *skew-symmetry* in the indices of $\mathbb{n}_{\alpha\beta} = \mathbb{n}_{[\alpha\beta]}$ and $\mathbb{N}_{\alpha\beta} = \mathbb{N}_{[\alpha\beta]}$.

In the following, corollaries will be used to assemble helpful intermediate results. For the sake of conciseness, these are explicitly outlined only for spatial quantities but hold likewise for material quantities with obvious adaption of the notation to upper case letters.

Corollary 2.2 (i). *Spatial atomistic position-wise sensitivities*

$$\partial\mathbb{x}_{\alpha\beta}/\partial\mathbb{x}_\alpha = -\mathbb{i} \quad \textit{and} \quad \partial\mathbb{x}_{\alpha\beta}/\partial\mathbb{x}_\beta = \mathbb{i}.$$

Corollary 2.2 (ii). *Spatial atomistic pair reciprocity*

[1] The superposition of a spatial rigid body motion with a time-dependent rotation $\boldsymbol{R}(t) \in \mathrm{SO}(3)$ and translation $\boldsymbol{c}(t)$ renders the spatial atomistic position and thus the resulting pair length vector as

$$\mathbb{x}^*_\alpha(t) = \boldsymbol{R}(t)\cdot\mathbb{x}_\alpha + \boldsymbol{c}(t) \quad \text{and} \quad \mathbb{x}^*_\beta(t) = \boldsymbol{R}(t)\cdot\mathbb{x}_\beta + \boldsymbol{c}(t) \implies \mathbb{x}^*_{\alpha\beta}(t) = \boldsymbol{R}(t)\cdot\mathbb{x}_{\alpha\beta}.$$

Obviously, $|\mathbb{x}^*_{\alpha\beta}(t)| = |\mathbb{x}_{\alpha\beta}|$ remains invariant under superposed rigid body motions and is thus *objective*. Extending the group SO(3) to O(3) (i.e. including reflections) does not change the result, thus $|\mathbb{x}^*_{\alpha\beta}(t)| = |\mathbb{x}_{\alpha\beta}|$ remains likewise invariant under inversion of space and is thus also *parity symmetric*.

[2] Regarding terminology, we understand a vector as a directed distance that is characterised by its length and its direction, whereby the latter possesses an orientation and a sense of direction.

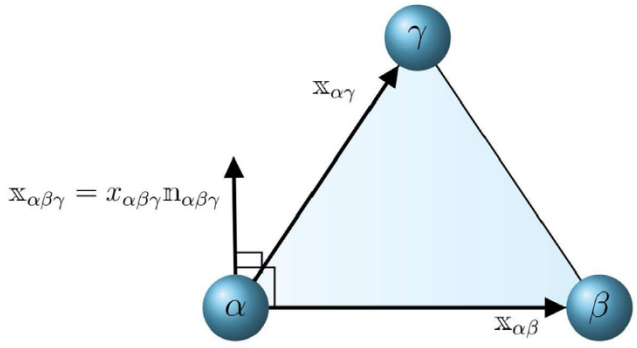


Fig. 3. Atomistic triplet area vector and triplet area.

The above sketch depicts the sign (sense of direction) of the pair length vector $\mathbb{x}_{\alpha\beta}$, whereby indices refer to vertices of an atomistic pair. Swapping the indices of $\mathbb{x}_{\alpha\beta}$ changes its sign, thus it is skew-symmetric in $\alpha\beta$, i.e. $\mathbb{x}_{\alpha\beta} \equiv \mathbb{x}_{[\alpha\beta]}$. Following the arrow for the sequence $\alpha\beta$ results in a positive sign, reversing the sequence to $\beta\alpha$ renders a negative sign.

$$\mathbb{x}_{\alpha\beta} \equiv -\mathbb{x}_{\beta\alpha} \implies \mathbb{n}_{\alpha\beta} \equiv -\mathbb{n}_{\beta\alpha} \quad \textit{with} \quad x_{\alpha\beta} \equiv x_{\beta\alpha}.$$

Corollary 2.2 (iii). *Spatial atomistic pair-wise sensitivities*

$$\partial x_{\alpha\beta}/\partial \mathbb{x}_{\alpha\beta} = \mathbb{n}_{\alpha\beta}.$$

Spatial atomistic pair-wise sensitivity: fully symmetric pair-wise orthogonal projection tensor

$$\partial \mathbb{n}_{\alpha\beta}/\partial \mathbb{x}_{\alpha\beta} = [\mathbb{i} - \mathbb{n}_{\alpha\beta} \otimes \mathbb{n}_{\alpha\beta}]/x_{\alpha\beta} := \mathbb{p}^{\perp}_{\alpha\beta}.$$

2.2.2. Pair stretches

Inspired by continuum kinematics, we also introduce spatial and material *pair stretches* as

$$\lambda_{\alpha\beta} := x_{\alpha\beta}/X_{\alpha\beta} \quad \text{and} \quad \Lambda_{\alpha\beta} := X_{\alpha\beta}/x_{\alpha\beta}. \tag{5}$$

Note the *symmetry* in the indices of $\lambda_{\alpha\beta} = \lambda_{(\alpha\beta)}$ and $\Lambda_{\alpha\beta} = \Lambda_{(\alpha\beta)}$, reflecting that the spatial and material pair stretches associated with each atom belonging to a specific atomistic pair are the same.

It is trivially established from the preceding discussion on invariance that the pair stretches are objective and parity symmetric.

Corollary 2.2 (iv). *Spatial atomistic pair-wise and position-wise sensitivities*

$$\partial \lambda_{\alpha\beta}/\partial \mathbb{x}_{\alpha\beta} = \mathbb{n}_{\alpha\beta}/X_{\alpha\beta} \quad \textit{and} \quad \partial \lambda_{\alpha\beta}/\partial \mathbb{x}_{\alpha} = -\mathbb{n}_{\alpha\beta}/X_{\alpha\beta}.$$

2.3. Atomistic triplets

2.3.1. Triplet areas

Next, we introduce the spatial and material *triplet area vectors* pointing perpendicular to the plane spanned by the atomistic pairs $\alpha\beta \to \alpha\gamma$, see Fig. 3, as

$$\begin{aligned} \mathbb{x}_{\alpha\beta\gamma} := \mathbb{x}_{\alpha\beta} \times \mathbb{x}_{\alpha\gamma} &=: \widehat{\mathbb{x}}_{\alpha\beta} \cdot \mathbb{x}_{\alpha\gamma} \\ &=: \mathbb{x}_{\alpha\beta} \cdot \widehat{\mathbb{x}}_{\alpha\gamma} \end{aligned} \tag{6}$$

and

$$\begin{aligned} \mathbb{X}_{\alpha\beta\gamma} := \mathbb{X}_{\alpha\beta} \times \mathbb{X}_{\alpha\gamma} &=: \widehat{\mathbb{X}}_{\alpha\beta} \cdot \mathbb{X}_{\alpha\gamma} \\ &=: \mathbb{X}_{\alpha\beta} \cdot \widehat{\mathbb{X}}_{\alpha\gamma} \end{aligned}$$

Here, $\widehat{\{\bullet\}}$ denotes the skew-symmetric (spin) tensor homoeomorphic to the (axial) vector $\{\bullet\}$. Note the minor *skew-symmetry* in the second pair of indices of $\mathbb{x}_{\alpha\beta\gamma}$ and $\mathbb{X}_{\alpha\beta\gamma}$ that results immediately from the skew-symmetric properties of the vector product. Moreover, due to the invariance of the vector-valued area of the enclosed triplet, $\mathbb{x}_{\alpha\beta\gamma}$ and $\mathbb{X}_{\alpha\beta\gamma}$ allow for cyclic permutations of $\alpha\beta\gamma$. Taken together, $\mathbb{x}_{\alpha\beta\gamma} = \mathbb{x}_{[\alpha\beta\gamma]}$ and $\mathbb{X}_{\alpha\beta\gamma} = \mathbb{X}_{[\alpha\beta\gamma]}$ are *fully skew-symmetric* in $\alpha\beta\gamma$ (cf. the skew-symmetries of the third-order Levi-Civita symbol).

In line with the requirement of invariance,[3] it is convenient to introduce the corresponding scalar-valued spatial and material *triplet areas* as

$$x_{\alpha\beta\gamma} := |\mathbb{x}_{\alpha\beta\gamma}| \quad \text{and} \quad X_{\alpha\beta\gamma} := |\mathbb{X}_{\alpha\beta\gamma}|. \tag{7}$$

Note the *full symmetry* in the indices of $x_{\alpha\beta\gamma} = x_{(\alpha\beta\gamma)}$ and $X_{\alpha\beta\gamma} = X_{(\alpha\beta\gamma)}$, reflecting that the spatial and material triplet areas associated with each atom belonging to a specific atomistic triplet are the same. Consequently, the vector-valued spatial and material *triplet directions* (triplet normals) follow as

$$\mathbb{n}_{\alpha\beta\gamma} := \mathbb{x}_{\alpha\beta\gamma}/x_{\alpha\beta\gamma} \quad \text{and} \quad \mathbb{N}_{\alpha\beta\gamma} := \mathbb{X}_{\alpha\beta\gamma}/X_{\alpha\beta\gamma}. \tag{8}$$

Note the *full skew-symmetry* in the indices of $\mathbb{n}_{\alpha\beta\gamma} = \mathbb{n}_{[\alpha\beta\gamma]}$ and $\mathbb{N}_{\alpha\beta\gamma} = \mathbb{N}_{[\alpha\beta\gamma]}$.

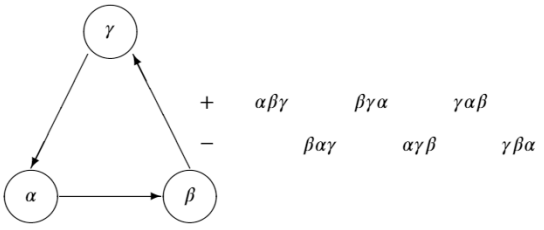
Corollary 2.3 (i). *Spatial atomistic pair-wise sensitivities*

$$\partial\mathbb{x}_{\alpha\beta\gamma}/\partial\mathbb{x}_{\alpha\beta} = -\widehat{\mathbb{x}}_{\alpha\gamma} \quad \textit{and} \quad \partial\mathbb{x}_{\alpha\beta\gamma}/\partial\mathbb{x}_{\alpha\gamma} = \widehat{\mathbb{x}}_{\alpha\beta}.$$

Spatial atomistic position-wise sensitivities

$$\partial\mathbb{x}_{\alpha\beta\gamma}/\partial\mathbb{x}_{\alpha} = \widehat{\mathbb{x}}_{\alpha\gamma} - \widehat{\mathbb{x}}_{\alpha\beta} = \widehat{\mathbb{x}}_{\beta\gamma} \quad \textit{and} \quad \partial\mathbb{x}_{\alpha\beta\gamma}/\partial\mathbb{x}_{\beta} = -\widehat{\mathbb{x}}_{\alpha\gamma} \quad \textit{and} \quad \partial\mathbb{x}_{\alpha\beta\gamma}/\partial\mathbb{x}_{\gamma} = \widehat{\mathbb{x}}_{\alpha\beta}.$$

Corollary 2.3 (ii). *Spatial atomistic triplet reciprocity*



The above sketch depicts the sign (sence of direction) of the triplet area vector $\mathbb{x}_{\alpha\beta\gamma}$, whereby indices refer to vertices of an atomistic triplet. Swapping any two indices of $\mathbb{x}_{\alpha\beta\gamma}$ changes its sign, thus it is fully skew-symmetric in $\alpha\beta\gamma$, i.e. $\mathbb{x}_{\alpha\beta\gamma} \equiv \mathbb{x}_{[\alpha\beta\gamma]}$. Following the arrows for the sequence $\alpha\beta\gamma$ results in a positive sign, changing the orientation for the circuit in the $\alpha\beta\gamma$ renders a negative sign.

$$\begin{aligned}
&\mathbb{x}_{\alpha\beta\gamma} \equiv \mathbb{x}_{\beta\gamma\alpha} \equiv \mathbb{x}_{\gamma\alpha\beta} \equiv -\mathbb{x}_{\beta\alpha\gamma} \equiv -\mathbb{x}_{\alpha\gamma\beta} \equiv -\mathbb{x}_{\gamma\beta\alpha} \quad \Longrightarrow \\
&\mathbb{n}_{\alpha\beta\gamma} \equiv \mathbb{n}_{\beta\gamma\alpha} \equiv \mathbb{n}_{\gamma\alpha\beta} \equiv -\mathbb{n}_{\beta\alpha\gamma} \equiv -\mathbb{n}_{\alpha\gamma\beta} \equiv -\mathbb{n}_{\gamma\beta\alpha} \quad \textit{with} \\
&x_{\alpha\beta\gamma} \equiv x_{\beta\gamma\alpha} \equiv x_{\gamma\alpha\beta} \equiv \quad x_{\beta\alpha\gamma} \equiv \quad x_{\alpha\gamma\beta} \equiv \quad x_{\gamma\beta\alpha}
\end{aligned}$$

Corollary 2.3 (iii). *Spatial atomistic triplet-wise sensitivities*

$$\partial x_{\alpha\beta\gamma}/\partial\mathbb{x}_{\alpha\beta\gamma} = \mathbb{n}_{\alpha\beta\gamma}.$$

Spatial atomistic triplet-wise sensitivity: fully symmetric triplet-wise orthogonal projection tensor

$$\partial\mathbb{n}_{\alpha\beta\gamma}/\partial\mathbb{x}_{\alpha\beta\gamma} = [\mathbb{i} - \mathbb{n}_{\alpha\beta\gamma} \otimes \mathbb{n}_{\alpha\beta\gamma}]/x_{\alpha\beta\gamma} := \mathbb{p}^{\perp}_{\alpha\beta\gamma}.$$

2.3.2. Triplet stretches

Again inspired by continuum kinematics, we introduce, as a novel concept, spatial and material *triplet stretches* as

$$\lambda_{\alpha\beta\gamma} := x_{\alpha\beta\gamma}/X_{\alpha\beta\gamma} \quad \text{and} \quad \Lambda_{\alpha\beta\gamma} := X_{\alpha\beta\gamma}/x_{\alpha\beta\gamma}. \tag{9}$$

Note the *full symmetry* in the indices of $\lambda_{\alpha\beta\gamma} = \lambda_{(\alpha\beta\gamma)}$ and $\Lambda_{\alpha\beta\gamma} = \Lambda_{(\alpha\beta\gamma)}$, reflecting that the spatial and material triplet stretches associated with each atom belonging to a specific atomistic triplet are the same.

It is trivially established from the preceding discussion on invariance that the triplet stretches are objective and parity symmetric.

Corollary 2.3 (iv). *Spatial atomistic triplet-wise and position-wise sensitivities*

$$\partial\lambda_{\alpha\beta\gamma}/\partial\mathbb{x}_{\alpha\beta\gamma} = \mathbb{n}_{\alpha\beta\gamma}/X_{\alpha\beta\gamma} \quad \textit{and} \quad \partial\lambda_{\alpha\beta\gamma}/\partial\mathbb{x}_{\alpha} = -\mathbb{x}_{\beta\gamma} \times \mathbb{n}_{\alpha\beta\gamma}/X_{\alpha\beta\gamma}.$$

[3] The superposition of a spatial rigid body motion with a time-dependent rotation $\boldsymbol{R}(t) \in \mathrm{SO}(3)$ (and a translation $\boldsymbol{c}(t)$) renders the spatial pair length vectors and thus the resulting triplet area vector as

$$\mathbb{x}^{*}_{\alpha\beta}(t) = \boldsymbol{R}(t) \cdot \mathbb{x}_{\alpha\beta} \quad \text{and} \quad \mathbb{x}^{*}_{\alpha\gamma}(t) = \boldsymbol{R}(t) \cdot \mathbb{x}_{\alpha\gamma} \implies \mathbb{x}^{*}_{\alpha\beta\gamma}(t) = \det \boldsymbol{R}(t)\, \boldsymbol{R}(t) \cdot \mathbb{x}_{\alpha\beta\gamma}.$$

Obviously, $|\mathbb{x}^{*}_{\alpha\beta\gamma}(t)| = |\mathbb{x}_{\alpha\beta\gamma}|$ remains invariant under superposed rigid body motions and is thus objective. Extending the group SO(3) to O(3) does not change the result, thus $|\mathbb{x}^{*}_{\alpha\beta\gamma}(t)| = |\mathbb{x}_{\alpha\beta\gamma}|$ remains likewise invariant under inversion of space and is thus also parity symmetric.

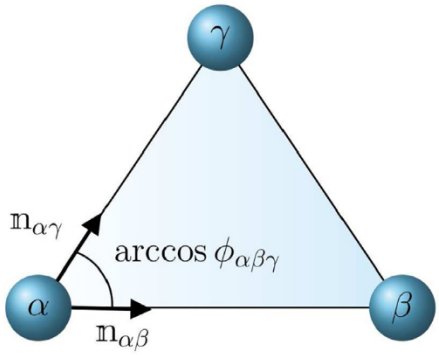


Fig. 4. Atomistic triplet angle.

2.3.3. Triplet angles

Classically, the intuitive concept of rotational springs in terms of angles between adjacent atomistic pairs motivates the introduction of spatial and material *triplet angles*, see Fig. 4. However, here, for convenience, we uniquely re-assign triplet angles from the interval $[0,\pi]$ to the interval $[+1,-1]$ by the cosine map (still denoting the result, admittedly somewhat sloppily, as triplet angles), thus

$$\begin{aligned}\phi_{\alpha\beta\gamma} &:= \mathbb{m}_{\alpha\beta}\cdot\mathbb{m}_{\alpha\gamma}\\ &\equiv \mathbb{m}_{\alpha\gamma}\cdot\mathbb{m}_{\alpha\beta} =: \phi_{\alpha\gamma\beta}\end{aligned} \tag{10}$$

and

$$\begin{aligned}\boldsymbol{\Phi}_{\alpha\beta\gamma} &:= \mathbb{N}_{\alpha\beta}\cdot\mathbb{N}_{\alpha\gamma}\\ &\equiv \mathbb{N}_{\alpha\gamma}\cdot\mathbb{N}_{\alpha\beta} =: \boldsymbol{\Phi}_{\alpha\gamma\beta}.\end{aligned}$$

Note the *minor symmetry* in the right pair of indices of $\phi_{\alpha\beta\gamma}=\phi_{\alpha(\beta\gamma)}$ and $\boldsymbol{\Phi}_{\alpha\beta\gamma}=\boldsymbol{\Phi}_{\alpha(\beta\gamma)}$, reflecting the commutativity of the scalar product. Yet, notably, the spatial and material triplet angles associated with each atom belonging to a specific atomistic triplet are in general distinct.

It is trivially established from the preceding discussion on invariance that the triplet angles are objective and parity symmetric.

Corollary 2.3 (v). *Spatial atomistic pair-wise sensitivities*

$$\partial\phi_{\alpha\beta\gamma}/\partial\mathbb{x}_{\alpha\beta}=\mathbb{p}^{\perp}_{\alpha\beta}\cdot\mathbb{m}_{\alpha\gamma}\quad\textit{and}\quad\partial\phi_{\alpha\beta\gamma}/\partial\mathbb{x}_{\alpha\gamma}=\mathbb{p}^{\perp}_{\alpha\gamma}\cdot\mathbb{m}_{\alpha\beta}.$$

Spatial atomistic position-wise sensitivities

$$\partial\phi_{\alpha\beta\gamma}/\partial\mathbb{x}_{\alpha}=-[\mathbb{p}^{\perp}_{\alpha\beta}\cdot\mathbb{m}_{\alpha\gamma}+\mathbb{p}^{\perp}_{\alpha\gamma}\cdot\mathbb{m}_{\alpha\beta}]\quad\textit{and}\quad\partial\phi_{\alpha\beta\gamma}/\partial\mathbb{x}_{\beta}=\mathbb{p}^{\perp}_{\alpha\beta}\cdot\mathbb{m}_{\alpha\gamma}\quad\textit{and}\quad\partial\phi_{\alpha\beta\gamma}/\partial\mathbb{x}_{\gamma}=\mathbb{p}^{\perp}_{\alpha\gamma}\cdot\mathbb{m}_{\alpha\beta}.$$

2.3.4. Triplet twists

In analogy to the triplet stretches, we also introduce, again as a novel concept, spatial and material *triplet twists* as

$$\omega_{\alpha\beta\gamma}:=\phi_{\alpha\beta\gamma}/\boldsymbol{\Phi}_{\alpha\beta\gamma}\quad\text{and}\quad\Omega_{\alpha\beta\gamma}:=\boldsymbol{\Phi}_{\alpha\beta\gamma}/\phi_{\alpha\beta\gamma}. \tag{11}$$

Note the *minor symmetry* in the right pair of indices of $\omega_{\alpha\beta\gamma}=\omega_{\alpha(\beta\gamma)}$ and $\Omega_{\alpha\beta\gamma}=\Omega_{\alpha(\beta\gamma)}$, reflecting the commutativity of the scalar product. Importantly, the spatial and material triplet twists associated with each atom belonging to a specific atomistic triplet are in general distinct.

It is trivially established from the preceding discussion on invariance that the triplet twists are objective and parity symmetric.

Corollary 2.3 (vi). *Spatial atomistic pair-wise sensitivities*

$$\partial\omega_{\alpha\beta\gamma}/\partial\mathbb{x}_{\alpha\beta}=\mathbb{p}^{\perp}_{\alpha\beta}\cdot\mathbb{m}_{\alpha\gamma}/\boldsymbol{\Phi}_{\alpha\beta\gamma}\quad\textit{and}\quad\partial\omega_{\alpha\beta\gamma}/\partial\mathbb{x}_{\alpha\gamma}=\mathbb{p}^{\perp}_{\alpha\gamma}\cdot\mathbb{m}_{\alpha\beta}/\boldsymbol{\Phi}_{\alpha\beta\gamma}$$

Spatial atomistic position-wise sensitivities

$$\partial\omega_{\alpha\beta\gamma}/\partial\mathbb{x}_{\alpha}=-[\mathbb{p}^{\perp}_{\alpha\beta}\cdot\mathbb{m}_{\alpha\gamma}+\mathbb{p}^{\perp}_{\alpha\gamma}\cdot\mathbb{m}_{\alpha\beta}]/\boldsymbol{\Phi}_{\alpha\beta\gamma}$$

$$\partial\omega_{\alpha\beta\gamma}/\partial\mathbb{x}_{\beta}=\mathbb{p}^{\perp}_{\alpha\beta}\cdot\mathbb{m}_{\alpha\gamma}/\boldsymbol{\Phi}_{\alpha\beta\gamma}\quad\textit{and}\quad\partial\omega_{\alpha\beta\gamma}/\partial\mathbb{x}_{\gamma}=\mathbb{p}^{\perp}_{\alpha\gamma}\cdot\mathbb{m}_{\alpha\beta}/\boldsymbol{\Phi}_{\alpha\beta\gamma}.$$

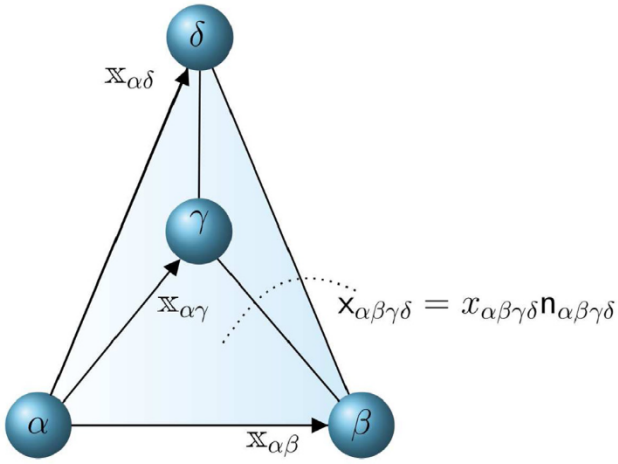


Fig. 5. Atomistic tetrad signed volume and tetrad volume.

2.4. Atomistic tetrads

2.4.1. Tetrad volumes

Finally, we introduce the spatial and material *tetrad signed volumes* contained within the atomistic pairs $\alpha\beta \to \alpha\gamma \to \alpha\delta$, see Fig. 5, as

$$\begin{aligned} \mathsf{x}_{\alpha\beta\gamma\delta} &:= [\mathbb{x}_{\alpha\beta} \times \mathbb{x}_{\alpha\gamma}] \cdot \mathbb{x}_{\alpha\delta} &&= \mathbb{x}_{\alpha\beta\gamma} \cdot \mathbb{x}_{\alpha\delta} \\ &&&= \mathbb{x}_{\alpha\delta\beta} \cdot \mathbb{x}_{\alpha\gamma} \quad \text{and} \\ &&&= \mathbb{x}_{\alpha\gamma\delta} \cdot \mathbb{x}_{\alpha\beta} \end{aligned} \qquad \begin{aligned} \mathsf{X}_{\alpha\beta\gamma\delta} &:= [\mathbb{X}_{\alpha\beta} \times \mathbb{X}_{\alpha\gamma}] \cdot \mathbb{X}_{\alpha\delta} &&= \mathbb{X}_{\alpha\beta\gamma} \cdot \mathbb{X}_{\alpha\delta} \\ &&&= \mathbb{X}_{\alpha\delta\beta} \cdot \mathbb{X}_{\alpha\gamma} \\ &&&= \mathbb{X}_{\alpha\gamma\delta} \cdot \mathbb{X}_{\alpha\beta} \end{aligned} \tag{12}$$

Note the full *skew-symmetry* in the last triplet of indices of $\mathsf{x}_{\alpha\beta\gamma\delta}$ and $\mathsf{X}_{\alpha\beta\gamma\delta}$ that results immediately from the skew-symmetric properties of the scalar triple product. Moreover, $\mathsf{x}_{\alpha\beta\gamma\delta}$ and $\mathsf{X}_{\alpha\beta\gamma\delta}$ are, due to the skew-symmetry properties of $\mathbb{x}_{\alpha\beta\gamma}$ and $\mathbb{X}_{\alpha\beta\gamma}$, also fully *skew-symmetric* in the first triplet of indices. Consequently, $\mathsf{x}_{\alpha\beta\gamma\delta} = \mathsf{x}_{[\alpha\beta\gamma\delta]}$ and $\mathsf{X}_{\alpha\beta\gamma\delta} = \mathsf{X}_{[\alpha\beta\gamma\delta]}$ are *fully skew-symmetric* in $\alpha\beta\gamma\delta$.

In line with the requirement of invariance,[4] it is convenient to introduce the corresponding scalar-valued spatial and material *tetrad volumes* as

$$x_{\alpha\beta\gamma\delta} := |\mathsf{x}_{\alpha\beta\gamma\delta}| \quad \text{and} \quad X_{\alpha\beta\gamma\delta} := |\mathsf{X}_{\alpha\beta\gamma\delta}|. \tag{13}$$

Note the *full symmetry* in the indices of $x_{\alpha\beta\gamma\delta} = x_{(\alpha\beta\gamma\delta)}$ and $X_{\alpha\beta\gamma\delta} = X_{(\alpha\beta\gamma\delta)}$, reflecting that the spatial and material tetrad volumes associated with each atom belonging to a specific atomistic tetrad are the same. Consequently, the scalar-valued spatial and material *tetrad "directions"* (tetrad normals=signs) follow as

$$\mathsf{n}_{\alpha\beta\gamma\delta} := \mathsf{x}_{\alpha\beta\gamma\delta}/x_{\alpha\beta\gamma\delta} \quad \text{and} \quad \mathsf{N}_{\alpha\beta\gamma\delta} := \mathsf{X}_{\alpha\beta\gamma\delta}/X_{\alpha\beta\gamma\delta}. \tag{14}$$

Note the *full skew-symmetry* in the indices of $\mathsf{n}_{\alpha\beta\gamma\delta} = \mathsf{n}_{[\alpha\beta\gamma\delta]}$ and $\mathsf{N}_{\alpha\beta\gamma\delta} = \mathsf{N}_{[\alpha\beta\gamma\delta]}$.

Corollary 2.4 (i). *Spatial atomistic triplet-wise sensitivities*

$$\partial\mathsf{x}_{\alpha\beta\gamma\delta}/\partial\mathbb{x}_{\alpha\beta\gamma} = \mathbb{x}_{\alpha\delta} \quad \textit{and} \quad \partial\mathsf{x}_{\alpha\beta\gamma\delta}/\partial\mathbb{x}_{\alpha\delta\beta} = \mathbb{x}_{\alpha\gamma} \quad \textit{and} \quad \partial\mathsf{x}_{\alpha\beta\gamma\delta}/\partial\mathbb{x}_{\alpha\gamma\delta} = \mathbb{x}_{\alpha\beta}.$$

Spatial atomistic pair-wise sensitivities

$$\partial\mathsf{x}_{\alpha\beta\gamma\delta}/\partial\mathbb{x}_{\alpha\delta} = \mathbb{x}_{\alpha\beta\gamma} \quad \textit{and} \quad \partial\mathsf{x}_{\alpha\beta\gamma\delta}/\partial\mathbb{x}_{\alpha\gamma} = \mathbb{x}_{\alpha\delta\beta} \quad \textit{and} \quad \partial\mathsf{x}_{\alpha\beta\gamma\delta}/\partial\mathbb{x}_{\alpha\beta} = \mathbb{x}_{\alpha\gamma\delta}.$$

Spatial atomistic position-wise sensitivities

$$\partial\mathsf{x}_{\alpha\beta\gamma\delta}/\partial\mathbb{x}_{\delta} = \mathbb{x}_{\alpha\beta\gamma} \quad \textit{and} \quad \partial\mathsf{x}_{\alpha\beta\gamma\delta}/\partial\mathbb{x}_{\gamma} = \mathbb{x}_{\alpha\delta\beta} \quad \textit{and} \quad \partial\mathsf{x}_{\alpha\beta\gamma\delta}/\partial\mathbb{x}_{\beta} = \mathbb{x}_{\alpha\gamma\delta},$$

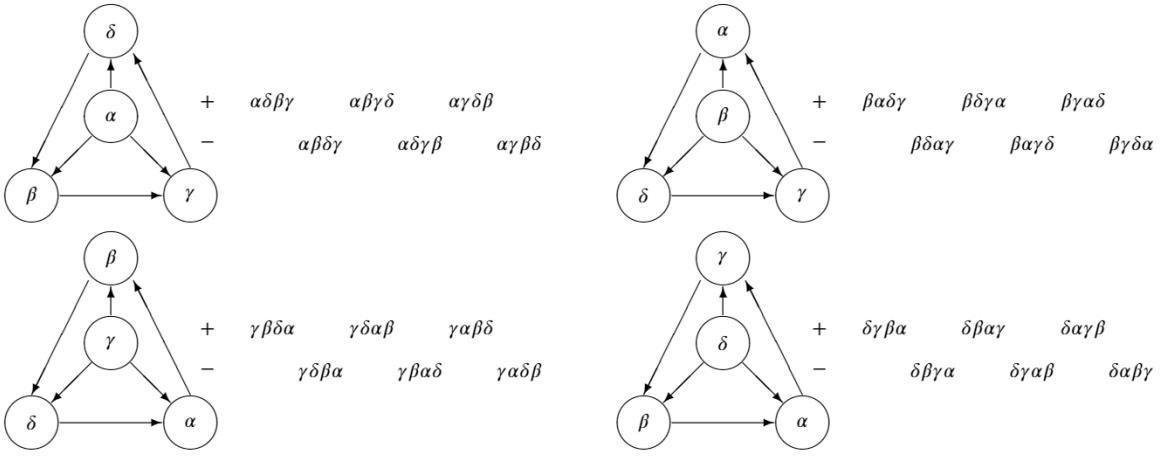
$$\partial\mathsf{x}_{\alpha\beta\gamma\delta}/\partial\mathbb{x}_{\alpha} = -\mathbb{x}_{\alpha\beta\gamma} - \mathbb{x}_{\alpha\delta\beta} - \mathbb{x}_{\alpha\gamma\delta} = \mathbb{x}_{\beta\delta\gamma}.$$

[4] The superposition of a spatial rigid body motion with a time-dependent rotation $\boldsymbol{R}(t) \in \mathrm{SO}(3)$ (and a translation $c(t)$) renders the spatial pair length vectors and thus the resulting tetrad signed volume as

$$\mathbb{x}^*_{\alpha\beta}(t) = \boldsymbol{R}(t) \cdot \mathbb{x}_{\alpha\beta} \quad \text{and} \quad \mathbb{x}^*_{\alpha\gamma}(t) = \boldsymbol{R}(t) \cdot \mathbb{x}_{\alpha\gamma} \quad \text{and} \quad \mathbb{x}^*_{\alpha\delta}(t) = \boldsymbol{R}(t) \cdot \mathbb{x}_{\alpha\delta} \implies \mathsf{x}^*_{\alpha\beta\gamma\delta}(t) = \det \boldsymbol{R}(t)\, \mathsf{x}_{\alpha\beta\gamma\delta}.$$

Obviously, $|\mathsf{x}^*_{\alpha\beta\gamma\delta}(t)| = |\mathsf{x}_{\alpha\beta\gamma\delta}|$ remains invariant under superposed rigid body motions and is thus objective. Extending the group SO(3) to O(3) does not change the result, thus $|\mathsf{x}^*_{\alpha\beta\gamma\delta}(t)| = |\mathsf{x}_{\alpha\beta\gamma\delta}|$ remains likewise invariant under inversion of space and is thus also parity symmetric.

Corollary 2.4 (ii). *Spatial atomistic tetrad reciprocity*



The above sketch depicts the sign of the tetrad signed volumes $\mathsf{x}_{\alpha\beta\gamma\delta}$*, whereby indices refer to vertices of an atomistic tetrad. Swapping any two indices of* $\mathsf{x}_{\alpha\beta\gamma\delta}$ *changes its sign, thus it is fully skew-symmetric in* $\alpha\beta\gamma\delta$*, i.e.* $\mathsf{x}_{\alpha\beta\gamma\delta} \equiv \mathsf{x}_{[\alpha\beta\gamma\delta]}$*. Following the arrows for the sequence* $\alpha\beta\gamma\delta$ *results in a positive sign, changing the orientation for the outer circuit in the* $\beta\gamma\delta$ *renders a negative sign.*

$$
\begin{aligned}
&\mathsf{x}_{\alpha\delta\beta\gamma} \equiv \mathsf{x}_{\alpha\beta\gamma\delta} \equiv \mathsf{x}_{\alpha\gamma\delta\beta} \equiv -\mathsf{x}_{\alpha\beta\delta\gamma} \equiv -\mathsf{x}_{\alpha\delta\gamma\beta} \equiv -\mathsf{x}_{\alpha\gamma\beta\delta} && \equiv\\
&\mathsf{x}_{\beta\alpha\delta\gamma} \equiv \mathsf{x}_{\beta\delta\gamma\alpha} \equiv \mathsf{x}_{\beta\gamma\alpha\delta} \equiv -\mathsf{x}_{\beta\delta\alpha\gamma} \equiv -\mathsf{x}_{\beta\alpha\gamma\delta} \equiv -\mathsf{x}_{\beta\gamma\delta\alpha} && \equiv\\
&\mathsf{x}_{\gamma\beta\delta\alpha} \equiv \mathsf{x}_{\gamma\delta\alpha\beta} \equiv \mathsf{x}_{\gamma\alpha\beta\delta} \equiv -\mathsf{x}_{\gamma\delta\beta\alpha} \equiv -\mathsf{x}_{\gamma\beta\alpha\delta} \equiv -\mathsf{x}_{\gamma\alpha\delta\beta} && \equiv\\
&\mathsf{x}_{\delta\gamma\beta\alpha} \equiv \mathsf{x}_{\delta\beta\alpha\gamma} \equiv \mathsf{x}_{\delta\alpha\gamma\beta} \equiv -\mathsf{x}_{\delta\beta\gamma\alpha} \equiv -\mathsf{x}_{\delta\gamma\alpha\beta} \equiv -\mathsf{x}_{\delta\alpha\beta\gamma} && \Longrightarrow\\
&\mathsf{n}_{\alpha\delta\beta\gamma} \equiv \mathsf{n}_{\alpha\beta\gamma\delta} \equiv \mathsf{n}_{\alpha\gamma\delta\beta} \equiv -\mathsf{n}_{\alpha\beta\delta\gamma} \equiv -\mathsf{n}_{\alpha\delta\gamma\beta} \equiv -\mathsf{n}_{\alpha\gamma\beta\delta} && \equiv\\
&\mathsf{n}_{\beta\alpha\delta\gamma} \equiv \mathsf{n}_{\beta\delta\gamma\alpha} \equiv \mathsf{n}_{\beta\gamma\alpha\delta} \equiv -\mathsf{n}_{\beta\delta\alpha\gamma} \equiv -\mathsf{n}_{\beta\alpha\gamma\delta} \equiv -\mathsf{n}_{\beta\gamma\delta\alpha} && \equiv\\
&\mathsf{n}_{\gamma\beta\delta\alpha} \equiv \mathsf{n}_{\gamma\delta\alpha\beta} \equiv \mathsf{n}_{\gamma\alpha\beta\delta} \equiv -\mathsf{n}_{\gamma\delta\beta\alpha} \equiv -\mathsf{n}_{\gamma\beta\alpha\delta} \equiv -\mathsf{n}_{\gamma\alpha\delta\beta} && \equiv\\
&\mathsf{n}_{\delta\gamma\beta\alpha} \equiv \mathsf{n}_{\delta\beta\alpha\gamma} \equiv \mathsf{n}_{\delta\alpha\gamma\beta} \equiv -\mathsf{n}_{\delta\beta\gamma\alpha} \equiv -\mathsf{n}_{\delta\gamma\alpha\beta} \equiv -\mathsf{n}_{\delta\alpha\beta\gamma} && \textit{with}\\
&x_{\alpha\delta\beta\gamma} \equiv x_{\alpha\beta\gamma\delta} \equiv x_{\alpha\gamma\delta\beta} \equiv \quad x_{\alpha\beta\delta\gamma} \equiv \quad x_{\alpha\delta\gamma\beta} \equiv \quad x_{\alpha\gamma\beta\delta} && \equiv\\
&x_{\beta\alpha\delta\gamma} \equiv x_{\beta\delta\gamma\alpha} \equiv x_{\beta\gamma\alpha\delta} \equiv \quad x_{\beta\delta\alpha\gamma} \equiv \quad x_{\beta\alpha\gamma\delta} \equiv \quad x_{\beta\gamma\delta\alpha} && \equiv\\
&x_{\gamma\beta\delta\alpha} \equiv x_{\gamma\delta\alpha\beta} \equiv x_{\gamma\alpha\beta\delta} \equiv \quad x_{\gamma\delta\beta\alpha} \equiv \quad x_{\gamma\beta\alpha\delta} \equiv \quad x_{\gamma\alpha\delta\beta} && \equiv\\
&x_{\delta\gamma\beta\alpha} \equiv x_{\delta\beta\alpha\gamma} \equiv x_{\delta\alpha\gamma\beta} \equiv \quad x_{\delta\beta\gamma\alpha} \equiv \quad x_{\delta\gamma\alpha\beta} \equiv \quad x_{\delta\alpha\beta\gamma}
\end{aligned}
$$

Corollary 2.4 (iii). *Spatial atomistic tetrad-wise sensitivities*

$$\partial x_{\alpha\beta\gamma\delta}/\partial \mathsf{x}_{\alpha\beta\gamma\delta} = \mathsf{n}_{\alpha\beta\gamma\delta}.$$

Spatial atomistic tetrad-wise sensitivity: fully symmetric tetrad-wise orthogonal projection tensor

$$\partial \mathsf{n}_{\alpha\beta\gamma\delta}/\partial \mathsf{x}_{\alpha\beta\gamma\delta} = [\mathbb{1} - \mathsf{n}_{\alpha\beta\gamma\delta} \otimes \mathsf{n}_{\alpha\beta\gamma\delta}]/x_{\alpha\beta\gamma\delta} := \mathsf{p}^{\perp}_{\alpha\beta\gamma\delta} \equiv 0.$$

2.4.2. Tetrad stretches

Again inspired by continuum kinematics, we also introduce, for the first time, spatial and material *tetrad stretches* as

$$\lambda_{\alpha\beta\gamma\delta} := x_{\alpha\beta\gamma\delta}/X_{\alpha\beta\gamma\delta} \quad \text{and} \quad \Lambda_{\alpha\beta\gamma\delta} := X_{\alpha\beta\gamma\delta}/x_{\alpha\beta\gamma\delta}. \tag{15}$$

Note the *full symmetry* in the indices of $\lambda_{\alpha\beta\gamma\delta} = \lambda_{(\alpha\beta\gamma\delta)}$ and $\Lambda_{\alpha\beta\gamma\delta} = \Lambda_{(\alpha\beta\gamma\delta)}$, reflecting that the spatial and material tetrad stretches associated with each atom belonging to a specific atomistic tetrad are the same.

It is trivially established from the preceding discussion on invariance that the tetrad stretches are objective and parity symmetric.

Corollary 2.4 (iv). *Spatial atomistic tetrad-wise and position-wise sensitivities*

$$\partial \lambda_{\alpha\beta\gamma\delta}/\partial \mathsf{x}_{\alpha\beta\gamma\delta} = \mathsf{n}_{\alpha\beta\gamma\delta}/X_{\alpha\beta\gamma\delta} \quad \textit{and} \quad \partial \lambda_{\alpha\beta\gamma\delta}/\partial \mathbb{x}_{\alpha} = -\mathbb{x}_{\beta\gamma\delta}\, \mathsf{n}_{\alpha\beta\gamma\delta}/X_{\alpha\beta\gamma\delta}.$$

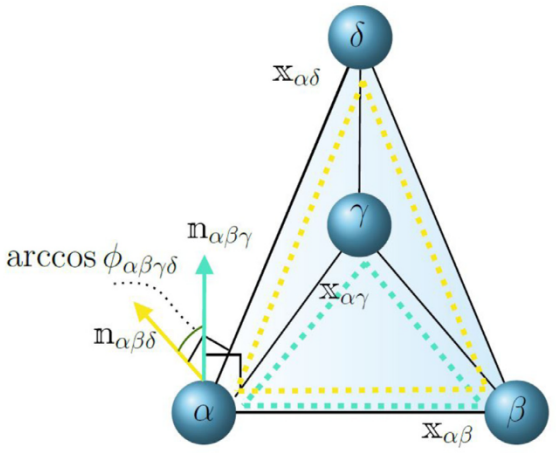


Fig. 6. Atomistic tetrad angle.

2.4.3. Tetrad dihedral angles

Traditionally, the intuitive concept of rotational springs in terms of dihedral angles between planes spanned by adjacent atomistic triplets motivates the introduction of spatial and material *tetrad dihedral angles*, see Fig. 6. However, here, for convenience, we uniquely re-assign tetrad dihedral angles from the interval $[0, \pi]$ to the interval $[+1, -1]$ by the cosine map (still denoting the result as tetrad dihedral angles), thus

$$\begin{aligned} \phi_{\alpha\beta\gamma\delta} &:= \mathbb{n}_{\alpha\beta\gamma} \cdot \mathbb{n}_{\alpha\beta\delta} \\ &\equiv \mathbb{n}_{\alpha\beta\delta} \cdot \mathbb{n}_{\alpha\beta\gamma} =: \phi_{\alpha\beta\delta\gamma} \end{aligned} \tag{16}$$

and

$$\begin{aligned} \boldsymbol{\Phi}_{\alpha\beta\gamma\delta} &:= \mathbb{N}_{\alpha\beta\gamma} \cdot \mathbb{N}_{\alpha\beta\delta} \\ &\equiv \mathbb{N}_{\alpha\beta\delta} \cdot \mathbb{N}_{\alpha\beta\gamma} =: \boldsymbol{\Phi}_{\alpha\beta\delta\gamma}. \end{aligned}$$

Note the *minor symmetry* in the left and right pair of indices of $\phi_{\alpha\beta\gamma\delta} = \phi_{(\alpha\beta)\gamma\delta} = \phi_{\alpha\beta(\gamma\delta)}$ and $\boldsymbol{\Phi}_{\alpha\beta\gamma\delta} = \boldsymbol{\Phi}_{(\alpha\beta)\gamma\delta} = \boldsymbol{\Phi}_{\alpha\beta(\gamma\delta)}$, reflecting the properties of the triplet directions and the commutativity of the scalar product. Despite this, the spatial and material tetrad dihedral angles associated with each atom belonging to a specific atomistic tetrad are in general distinct.

It is trivially established from the preceding discussion on invariance that the tetrad dihedral angles are objective and parity symmetric.

Corollary 2.4 (v). *Spatial atomistic triplet-wise sensitivities*

$$\partial\phi_{\alpha\beta\gamma\delta}/\partial\mathbb{x}_{\alpha\beta\gamma} = \mathbb{p}^{\perp}_{\alpha\beta\gamma} \cdot \mathbb{n}_{\alpha\beta\delta} \quad \textit{and} \quad \partial\phi_{\alpha\beta\gamma\delta}/\partial\mathbb{x}_{\alpha\beta\delta} = \mathbb{p}^{\perp}_{\alpha\beta\delta} \cdot \mathbb{n}_{\alpha\beta\gamma}.$$

Spatial atomistic position-wise sensitivities

$$\begin{aligned} \partial\phi_{\alpha\beta\gamma\delta}/\partial\mathbb{x}_{\alpha} &= -\big[\mathbb{x}_{\beta\gamma} \times [\mathbb{p}^{\perp}_{\alpha\beta\gamma} \cdot \mathbb{n}_{\alpha\beta\delta}] + \mathbb{x}_{\beta\delta} \times [\mathbb{p}^{\perp}_{\alpha\beta\delta} \cdot \mathbb{n}_{\alpha\beta\gamma}]\big] \\ \partial\phi_{\alpha\beta\gamma\delta}/\partial\mathbb{x}_{\beta} &= +\big[\mathbb{x}_{\alpha\gamma} \times [\mathbb{p}^{\perp}_{\alpha\beta\gamma} \cdot \mathbb{n}_{\alpha\beta\delta}] + \mathbb{x}_{\alpha\delta} \times [\mathbb{p}^{\perp}_{\alpha\beta\delta} \cdot \mathbb{n}_{\alpha\beta\gamma}]\big] \\ \partial\phi_{\alpha\beta\gamma\delta}/\partial\mathbb{x}_{\gamma} &= -\big[\mathbb{x}_{\alpha\beta} \times [\mathbb{p}^{\perp}_{\alpha\beta\gamma} \cdot \mathbb{n}_{\alpha\beta\delta}]\big] \\ \partial\phi_{\alpha\beta\gamma\delta}/\partial\mathbb{x}_{\delta} &= -\big[\mathbb{x}_{\alpha\beta} \times [\mathbb{p}^{\perp}_{\alpha\beta\delta} \cdot \mathbb{n}_{\alpha\beta\gamma}]\big] \end{aligned}$$

2.4.4. Tetrad dihedral twists

Finally, in analogy to the tetrad stretches, we also introduce spatial and material *tetrad dihedral twists* as

$$\omega_{\alpha\beta\gamma\delta} := \phi_{\alpha\beta\gamma\delta}/\boldsymbol{\Phi}_{\alpha\beta\gamma\delta} \quad \text{and} \quad \Omega_{\alpha\beta\gamma\delta} := \boldsymbol{\Phi}_{\alpha\beta\gamma\delta}/\phi_{\alpha\beta\gamma\delta}. \tag{17}$$

Note the *minor symmetry* in the left and right pair of indices of $\omega_{\alpha\beta\gamma\delta} = \omega_{(\alpha\beta)\gamma\delta} = \omega_{\alpha\beta(\gamma\delta)}$ and $\Omega_{\alpha\beta\gamma\delta} = \Omega_{(\alpha\beta)\gamma\delta} = \Omega_{\alpha\beta(\gamma\delta)}$, reflecting the properties of the triplet directions and the commutativity of the scalar product. Despite possessing only minor symmetries, the spatial and material tetrad dihedral twists associated with each atom belonging to a specific atomistic tetrad are in general distinct.

It is trivially established from the preceding discussion on invariance that the tetrad dihedral twists are objective and parity symmetric.

Corollary 2.4 (vi). *Spatial atomistic triplet-wise sensitivities*

$$\partial\omega_{\alpha\beta\gamma\delta}/\partial\mathbb{x}_{\alpha\beta\gamma} = \mathbb{p}^{\perp}_{\alpha\beta\gamma} \cdot \mathbb{n}_{\alpha\beta\delta}/\boldsymbol{\Phi}_{\alpha\beta\gamma\delta} \quad \textit{and} \quad \partial\omega_{\alpha\beta\gamma\delta}/\partial\mathbb{x}_{\alpha\beta\delta} = \mathbb{p}^{\perp}_{\alpha\beta\delta} \cdot \mathbb{n}_{\alpha\beta\gamma}/\boldsymbol{\Phi}_{\alpha\beta\gamma\delta}.$$

Table 2
Summary of atomistic deformational and configurational mechanics.

Total potential energy		
$E := E^{\text{int}} + E^{\text{ext}}$		
$E^{\text{int}} := E^{\text{pair}} + E^{\text{triplet}} + E^{\text{tetrad}}$, $E^{\text{ext}} := \sum_\alpha V^\alpha(\mathbb{x}_\alpha)$		
Equilibrium of spatial atomistic forces		
$\mathrm{D}_\delta E = 0 \quad \forall \{\mathrm{D}_\delta \mathbb{x}_\epsilon\}$		
$\mathbb{k}^{\text{int}}_\alpha + \mathbb{k}^{\text{ext}}_\alpha \doteq \mathbb{0}$ with $\mathbb{k}^{\text{int}}_\alpha := \mathbb{k}^{\text{pair}}_\alpha + \mathbb{k}^{\text{triplet}}_\alpha + \mathbb{k}^{\text{tetrad}}_\alpha$ and $\mathbb{k}^{\text{ext}}_\alpha := -\partial V^{\text{ext}}_\alpha / \partial \mathbb{x}_\alpha$		
Atomistic pairs	Atomistic triplets	Atomistic tetrads
$\mathbb{k}^{\text{pair}}_\alpha := -\partial E^{\text{pair}} / \partial \mathbb{x}_\alpha$	$\mathbb{k}^{\text{triplet}}_\alpha := -\partial E^{\text{triplet}} / \partial \mathbb{x}_\alpha$	$\mathbb{k}^{\text{tetrad}}_\alpha := -\partial E^{\text{tetrad}} / \partial \mathbb{x}_\alpha$
Non-equilibrium of material atomistic forces		
$\mathrm{d}_\delta E := R_\delta \leq 0 \quad \forall \{\mathrm{d}_\delta \mathbb{X}_\epsilon\}$		
$\mathbb{K}^{\text{int}}_\alpha =: \mathbb{K}^{\text{mat}}_\alpha$ with $\mathbb{K}^{\text{int}}_\alpha := \mathbb{K}^{\text{pair}}_\alpha + \mathbb{K}^{\text{triplet}}_\alpha + \mathbb{K}^{\text{tetrad}}_\alpha$ and $R_\delta =: \sum_\alpha \mathbb{K}^{\text{mat}}_\alpha \cdot \mathrm{d}_\delta \mathbb{X}_\alpha$		
Atomistic pairs	Atomistic triplets	Atomistic tetrads
$\mathbb{K}^{\text{pair}}_\alpha := -\partial E^{\text{pair}} / \partial \mathbb{X}_\alpha$	$\mathbb{K}^{\text{triplet}}_\alpha := -\partial E^{\text{triplet}} / \partial \mathbb{X}_\alpha$	$\mathbb{K}^{\text{tetrad}}_\alpha := -\partial E^{\text{tetrad}} / \partial \mathbb{X}_\alpha$

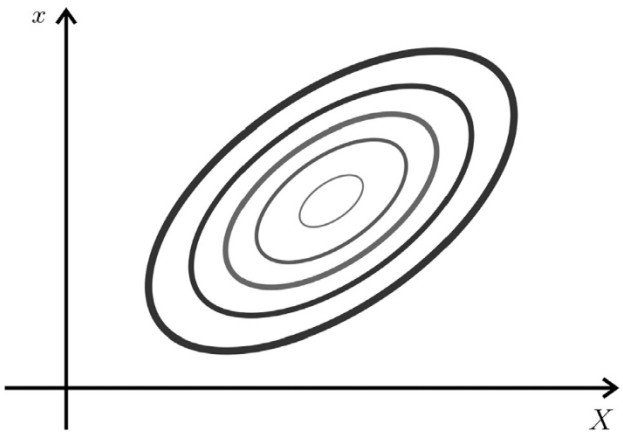


Fig. 7. Energy landscape $E(x, X)$ of a one-dof atomistic system depending on spatial and material positions.

Spatial atomistic position-wise sensitivities

$$
\begin{aligned}
\partial \omega_{\alpha\beta\gamma\delta} / \partial \mathbb{x}_\alpha &= -\big[\mathbb{x}_{\beta\gamma} \times [\mathbb{p}^{\perp}_{\alpha\beta\gamma} \cdot \mathbb{n}_{\alpha\beta\delta}] + \mathbb{x}_{\beta\delta} \times [\mathbb{p}^{\perp}_{\alpha\beta\delta} \cdot \mathbb{n}_{\alpha\beta\gamma}]\big] / \boldsymbol{\Phi}_{\alpha\beta\gamma\delta} \\
\partial \omega_{\alpha\beta\gamma\delta} / \partial \mathbb{x}_\beta &= +\big[\mathbb{x}_{\alpha\gamma} \times [\mathbb{p}^{\perp}_{\alpha\beta\gamma} \cdot \mathbb{n}_{\alpha\beta\delta}] + \mathbb{x}_{\alpha\delta} \times [\mathbb{p}^{\perp}_{\alpha\beta\delta} \cdot \mathbb{n}_{\alpha\beta\gamma}]\big] / \boldsymbol{\Phi}_{\alpha\beta\gamma\delta} \\
\partial \omega_{\alpha\beta\gamma\delta} / \partial \mathbb{x}_\gamma &= -\big[\mathbb{x}_{\alpha\beta} \times [\mathbb{p}^{\perp}_{\alpha\beta\gamma} \cdot \mathbb{n}_{\alpha\beta\delta}] \big] / \boldsymbol{\Phi}_{\alpha\beta\gamma\delta} \\
\partial \omega_{\alpha\beta\gamma\delta} / \partial \mathbb{x}_\delta &= -\big[\mathbb{x}_{\alpha\beta} \times [\mathbb{p}^{\perp}_{\alpha\beta\delta} \cdot \mathbb{n}_{\alpha\beta\gamma}]\big] / \boldsymbol{\Phi}_{\alpha\beta\gamma\delta}
\end{aligned}
$$

3. Deformational versus configurational mechanics

The pertinent relations regarding atomistic deformational versus configurational mechanics as referred to in this section are assembled for the sake of overview in Table 2.

3.1. Total potential energy

The total potential energy E of an atomistic system depends on the sets of spatial and material positions $\{\mathbb{x}_\epsilon\}$ and $\{\mathbb{X}_\epsilon\}$, respectively, of all atoms, see the simplified single degree of freedom (dof) example in Fig. 7. For the deformational problem, $\{\mathbb{x}_\epsilon\}$ denote the unknown *variables*, whereas $\{\mathbb{X}_\epsilon\}$ serve as a given *parametrisation*. For the configurational problem these roles are reversed, i.e. $\{\mathbb{X}_\epsilon\}$ denote the variables, whereas $\{\mathbb{x}_\epsilon\}$ serve as parametrisation. The total potential energy E consists of an internal and an external contribution:

$$E := E^{\text{int}} + E^{\text{ext}}. \tag{18}$$

The external potential energy captures the interaction of the finite atomistic system with the (infinite) external world. It depends, through the atom-wise external potential energy V^α, merely on the spatial atomistic positions $\{\mathbb{x}_\epsilon\}$ of all atoms ϵ. That is,

$$E^{\text{ext}} := \sum_\alpha V^\alpha(\mathbb{x}_\alpha). \tag{19}$$

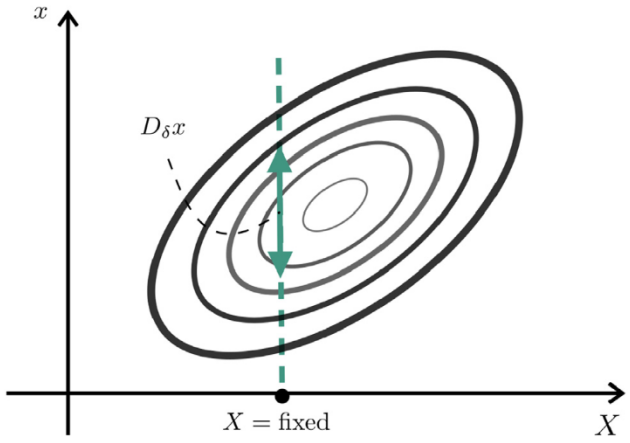


Fig. 8. Spatial variations in an energy landscape $E(x, X)$ of a one-dof atomistic system.

After condensing out the electronic degrees of freedom, the internal potential energy depends in general on the sets of spatial and material positions $\{\mathbb{x}_\epsilon\}$ and $\{\mathbb{X}_\epsilon\}$ of all atoms in the atomistic system and captures the interaction internal to the atomistic system. We shall here consider the special (however, often valid) assumption that an additive expansion into pair, triplet and tetrad potentials is sufficiently accurate to approximate the complex energetic landscape dictated by the underlying quantum mechanics, thus

$$E^{\text{int}} := E^{\text{pair}} + E^{\text{triplet}} + E^{\text{tetrad}}. \tag{20}$$

We will highlight the consequences of and various options for this expansion in the subsequent subsections.

3.2. Equilibrium of spatial atomistic forces

We shall here consider quasi-statics. Thus the equilibrium condition for spatial atomistic forces follows from *minimising* the total potential energy E of an atomistic system considered as a function of the spatial atomistic positions in $\{\mathbb{x}_\epsilon\}$ and parameterised in $\{\mathbb{X}_\epsilon\}$. We denote *spatial variations* of the spatial atomistic positions $\{\mathbb{x}_\epsilon\}$ at *fixed* material atomistic positions $\{\mathbb{X}_\epsilon\}$ as $\{\mathrm{D}_\delta\mathbb{x}_\epsilon\}$, see Fig. 8. Thus, the minimum condition for the total potential energy E under spatial variations reads

$$\mathrm{D}_\delta E = 0 \quad \forall\, \{\mathrm{D}_\delta\mathbb{x}_\epsilon\}. \tag{21}$$

Consequently, the minimum condition for the total potential energy E of an atomistic system is given by

$$\mathrm{D}_\delta E = -\sum_\alpha [\mathbb{k}_\alpha^{\text{int}} + \mathbb{k}_\alpha^{\text{ext}}] \cdot \mathrm{D}_\delta\mathbb{x}_\alpha := -\sum_\alpha [\mathbb{k}_\alpha^{\text{pair}} + \mathbb{k}_\alpha^{\text{triplet}} + \mathbb{k}_\alpha^{\text{tetrad}} + \mathbb{k}_\alpha^{\text{ext}}] \cdot \mathrm{D}_\delta\mathbb{x}_\alpha \doteq 0 \quad \forall\, \{\mathrm{D}_\delta\mathbb{x}_\epsilon\}. \tag{22}$$

In the above, $\mathbb{k}_\alpha^{\text{pair}}$, $\mathbb{k}_\alpha^{\text{triplet}}$ and $\mathbb{k}_\alpha^{\text{tetrad}}$ denote the contributions to the (net or rather *resultant*) internal spatial force acting on atom α that follow straightforwardly from the pair, triplet and tetrad potentials as

$$\mathbb{k}_\alpha^{\text{pair}} := -\frac{\partial E^{\text{pair}}}{\partial \mathbb{x}_\alpha}, \qquad \mathbb{k}_\alpha^{\text{triplet}} := -\frac{\partial E^{\text{triplet}}}{\partial \mathbb{x}_\alpha} \quad \text{and} \quad \mathbb{k}_\alpha^{\text{tetrad}} := -\frac{\partial E^{\text{tetrad}}}{\partial \mathbb{x}_\alpha}. \tag{23}$$

Likewise, the external spatial force acting on atom α is given explicitly by

$$\mathbb{k}_\alpha^{\text{ext}} := -\frac{\partial V_\alpha^{\text{ext}}}{\partial \mathbb{x}_\alpha}. \tag{24}$$

Since admissible spatial variations $\{\mathrm{D}_\delta\mathbb{x}_\epsilon\}$ of the spatial atomistic positions in $\{\mathbb{x}_\epsilon\}$ are *arbitrary*, the equilibrium condition for spatial forces acting on atom α results eventually as

$$\mathbb{k}_\alpha^{\text{int}} + \mathbb{k}_\alpha^{\text{ext}} \doteq \mathbb{0} \quad \text{with} \quad \mathbb{k}_\alpha^{\text{int}} := \mathbb{k}_\alpha^{\text{pair}} + \mathbb{k}_\alpha^{\text{triplet}} + \mathbb{k}_\alpha^{\text{tetrad}}. \tag{25}$$

Clearly, the above atomistic equilibrium condition is that the sum of all (resultant) internal spatial forces $\mathbb{k}_\alpha^{\text{int}} := \mathbb{k}_\alpha^{\text{pair}} + \mathbb{k}_\alpha^{\text{triplet}} + \mathbb{k}_\alpha^{\text{tetrad}}$ and external spatial forces $\mathbb{k}_\alpha^{\text{ext}}$ acting on atom α is zero.

3.3. Non-equilibrium of material atomistic forces

Here, we consider a *re-parametrisation* of the total potential energy E of an atomistic system as a function of the set of material atomistic positions $\{\mathbb{X}_\epsilon\}$, i.e. with $\{\mathbb{X}_\epsilon\}$ as the variables and $\{\mathbb{x}_\epsilon\}$ as the parametrisation. We denote *material variations* of the material

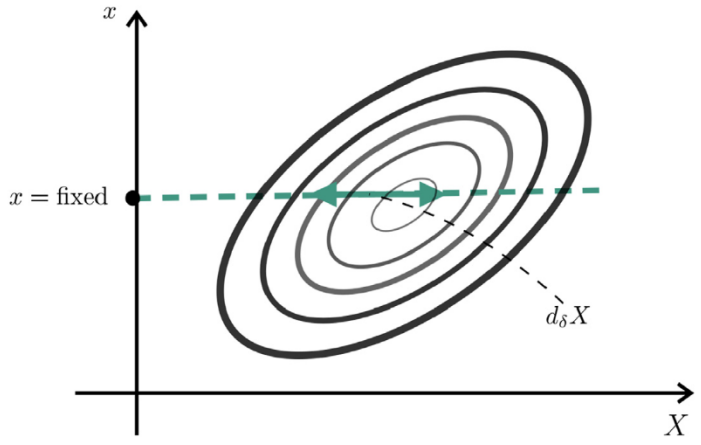


Fig. 9. Material variations in an energy landscape $E(x, X)$ of a one-dof atomistic system.

atomistic positions $\{\mathbb{X}_\epsilon\}$ at *fixed* spatial atomistic positions $\{\mathbb{x}_\epsilon\}$ as $\{\mathrm{d}_\delta\mathbb{X}_\epsilon\}$, see Fig. 9. Then the material variation of the total potential energy E equates with the *virtual energy release* R_δ that satisfies an inequality constraint in accordance with the second law, that is

$$\mathrm{d}_\delta E =: R_\delta \leq 0 \quad \forall\, \{\mathrm{d}_\delta\mathbb{X}_\epsilon\}. \tag{26}$$

Thereby, the material variation of the total potential energy E (note that $\mathrm{d}_\delta E^{\mathrm{ext}} \equiv 0$) is given by

$$\mathrm{d}_\delta E = -\sum_\alpha \mathbb{K}_\alpha^{\mathrm{int}} \cdot \mathrm{d}_\delta\mathbb{X}_\alpha := -\sum_\alpha [\mathbb{K}_\alpha^{\mathrm{pair}} + \mathbb{K}_\alpha^{\mathrm{triplet}} + \mathbb{K}_\alpha^{\mathrm{tetrad}}] \cdot \mathrm{d}_\delta\mathbb{X}_\alpha \quad \forall\, \{\mathrm{d}_\delta\mathbb{X}_\epsilon\}. \tag{27}$$

In the above, $\mathbb{K}_\alpha^{\mathrm{pair}}$, $\mathbb{K}_\alpha^{\mathrm{triplet}}$ and $\mathbb{K}_\alpha^{\mathrm{tetrad}}$ denote the contributions to the (resultant) internal material force acting on atom α that follow from the pair, triplet and tetrad potentials, respectively, as

$$\mathbb{K}_\alpha^{\mathrm{pair}} := -\frac{\partial E^{\mathrm{pair}}}{\partial \mathbb{X}_\alpha}, \qquad \mathbb{K}_\alpha^{\mathrm{triplet}} := -\frac{\partial E^{\mathrm{triplet}}}{\partial \mathbb{X}_\alpha} \quad \text{and} \quad \mathbb{K}_\alpha^{\mathrm{tetrad}} := -\frac{\partial E^{\mathrm{tetrad}}}{\partial \mathbb{X}_\alpha}. \tag{28}$$

Note that material variation of the total potential energy E does not in general render a stationary point (this would correspond to the rare case of configurational equilibrium) but defines a variation of total potential energy $\mathrm{d}_\delta E \neq 0$. A key question that follows from this observation is what are the energetic implications of a variation of the material configuration if we apply a deformation resulting in the same spatial configuration? According to the second law of thermodynamics, spontaneous configurational changes are only allowed if potential energy is *released*, thus $\mathrm{d}_\delta E \leq 0$. The corresponding virtual energy release $R_\delta \leq 0$ is then dissipated by other physical processes such as the propagation of cracks and/or defects. In accordance with analytical mechanics that associates energy variations with forces working on variations of kinematic quantities, we *define* the energy release R_δ as being due to material atomistic forces, that is

$$R_\delta =: \sum_\alpha \mathbb{K}_\alpha^{\mathrm{mat}} \cdot \mathrm{d}_\delta\mathbb{X}_\alpha \quad \forall\, \{\mathrm{d}_\delta\mathbb{X}_\epsilon\}. \tag{29}$$

According to the sign convention adopted, material variations $\mathrm{d}_\delta\mathbb{X}$ have to oppose material atomistic forces $\mathbb{K}_\alpha^{\mathrm{mat}}$ in order to result in a negative virtual energy release. Finally, since admissible material variations $\{\mathrm{d}_\delta\mathbb{X}_\epsilon\}$ of the material atomistic positions in $\{\mathbb{X}_\epsilon\}$ are arbitrary, the non-equilibrium condition for the material forces acting on atom α can be stated as

$$-\mathbb{K}_\alpha^{\mathrm{int}} =: \mathbb{K}_\alpha^{\mathrm{mat}} \quad \text{with} \quad \mathbb{K}_\alpha^{\mathrm{int}} := \mathbb{K}_\alpha^{\mathrm{pair}} + \mathbb{K}_\alpha^{\mathrm{triplet}} + \mathbb{K}_\alpha^{\mathrm{tetrad}}. \tag{30}$$

Taken together, the above atomistic non-equilibrium condition *defines* the negative sum of all (resultant) internal material forces $\mathbb{K}_\alpha^{\mathrm{int}} := \mathbb{K}_\alpha^{\mathrm{pair}} + \mathbb{K}_\alpha^{\mathrm{triplet}} + \mathbb{K}_\alpha^{\mathrm{tetrad}}$ acting on atom α as the material force $\mathbb{K}_\alpha^{\mathrm{mat}}$ energetically conjugate to material variations $\mathrm{d}_\delta\mathbb{X}_\alpha$ of the material position of atom α.

The above approach to deformational and configurational mechanics is elaborated in Fig. 10 for the simple paradigm of a spring with harmonic interaction potential.

4. Stretch-based energies

The pertinent relations regarding spatial and material stretch-based atomistic energies and interaction forces resulting therefrom that are referred to in this section are assembled for the sake of overview in Tables 3 and 4.

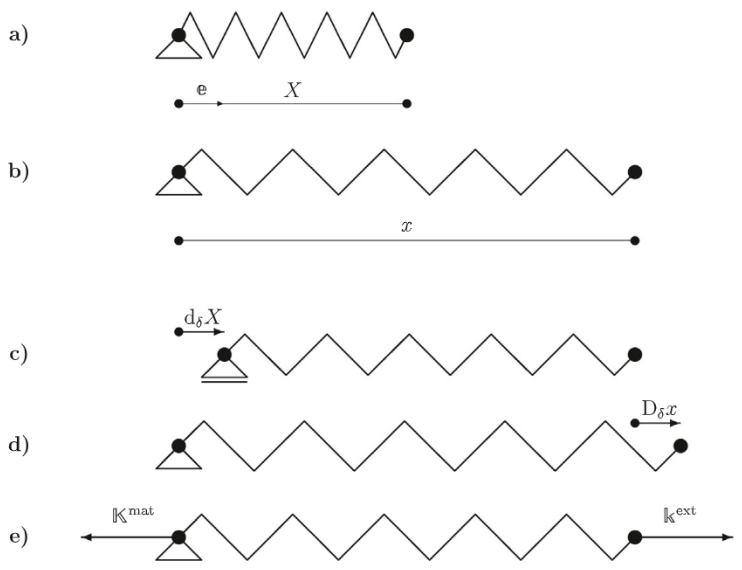


Fig. 10. Spring supported at the left end point and loaded by a given (spatial) force at the right end point: **(a)** material configuration with length X, **(b)** spatial configuration with length x, **(c)** material variation $\mathrm{d}_\delta X$, **(d)** spatial variation $\mathrm{D}_\delta x$, **(e)** applied spatial force $\mathbb{k}^{ext} := k^{ext}\mathbf{e}$ and resulting material force $\mathbb{K}^{mat} := K^{mat}\mathbf{e}$ (with $\mathbf{e}$ the horizontal base vector). For unit spring stiffness, the total potential energy reads $E = [x - X]^2/2 - k^{ext}x$, its spatial variation $\mathrm{D}_\delta E = [x - X - k^{ext}]\,\mathrm{D}_\delta x \doteq 0 \quad \forall\, \mathrm{D}_\delta x$ results in the equilibrium of spatial forces $k^{ext} = x - X$, its material variation $\mathrm{d}_\delta E = -[x - X]\,\mathrm{d}_\delta X := K^{mat}\,\mathrm{d}_\delta X \le 0 \quad \forall\, \mathrm{d}_\delta X$ determines the material force as $\mathbb{K}^{mat} = -\mathbb{k}^{ext}$, i.e. here as the reaction force at the support. If we imagine the support as a frictional slider, configurational changes, i.e. changes in the position of the support, occur once the material force reaches a given threshold $|K^{mat}| = K_0$. In a convex analysis setting the temporal evolution $\mathrm{d}_t X \propto -\mathrm{sign}\, K^{mat}$ follows here in the $\mathbf{e}$ direction.

4.1. Spatial stretch-based parametrisation

We consider expansions of the atomistic pair, triplet and tetrad contributions – E^{pair}, $E^{triplet}$ and E^{tetrad} – to the internal potential energy E^{int} in terms of their densities – $W_0^{\alpha\beta}$, $W_0^{\alpha\beta\gamma}$ and $W_0^{\alpha\beta\gamma\delta}$ – per unit *material* pair length $X_{\alpha\beta}$, triplet area $X_{\alpha\beta\gamma}$ and tetrad volume $X_{\alpha\beta\gamma\delta}$, respectively. We parameterise these energy densities in terms of spatial pair, triplet and tetrad stretches $\lambda_{\alpha\beta}$, $\lambda_{\alpha\beta\gamma}$ and $\lambda_{\alpha\beta\gamma\delta}$, respectively. Thus

$$E^{pair} := \frac{1}{2!}\sum_{\alpha,\beta} W_0^{\alpha\beta}(\lambda_{\alpha\beta})\, X_{\alpha\beta} \tag{31}$$

$$E^{triplet} := \frac{1}{3!}\sum_{\alpha,\beta,\gamma} W_0^{\alpha\beta\gamma}(\lambda_{\alpha\beta\gamma})\, X_{\alpha\beta\gamma} \tag{32}$$

$$E^{tetrad} := \frac{1}{4!}\sum_{\alpha,\beta,\gamma,\delta} W_0^{\alpha\beta\gamma\delta}(\lambda_{\alpha\beta\gamma\delta})\, X_{\alpha\beta\gamma\delta}. \tag{33}$$

Observe that, in contrast to the common approach adopted when defining atomistic pair potentials, we parameterise the pair potential (density) in terms of the spatial pair stretch rather than the spatial pair length.[5] The parametrisation of the triplet and tetrad potential densities in terms of the triplet and tetrad stretches follows the same philosophy. Recall that the spatial pair, triplet and tetrad stretches $\lambda_{\alpha\beta}$, $\lambda_{\alpha\beta\gamma}$ and $\lambda_{\alpha\beta\gamma\delta}$, respectively, are the same for all atoms belonging to a specific atomistic pair, triplet and tetrad, respectively. As a result their contribution is included only once in each summand of the above expansions.

[5] Traditionally, the Lennard-Jones two-body potential $\varphi^{\alpha\beta}$ expands in terms of the *spatial pair length* (distance between atomistic pairs) $x_{\alpha\beta}$ and two material parameters, the potential depth ϵ and the spatial pair length σ for which $\varphi^{\alpha\beta} \equiv 0$, as

$$\varphi^{\alpha\beta}(x_{\alpha\beta}) = 4\epsilon\left[\left[\frac{\sigma}{x_{\alpha\beta}}\right]^{12} - \left[\frac{\sigma}{x_{\alpha\beta}}\right]^{6}\right].$$

Reformulated as energy density $W_0^{\alpha\beta} := \varphi^{\alpha\beta}/X_{\alpha\beta}$ per unit material pair length $X_{\alpha\beta}$ and re-parameterised in terms of the *spatial pair stretch* $\lambda_{\alpha\beta}$, the Lennard-Jones potential expands alternatively as

$$W_0^{\alpha\beta}(\lambda_{\alpha\beta}) = 4\epsilon_0\left[\left[\frac{\sigma_0}{\lambda_{\alpha\beta}}\right]^{12} - \left[\frac{\sigma_0}{\lambda_{\alpha\beta}}\right]^{6}\right].$$

Here, we consider the re-defined material parameters $\epsilon_0 := \epsilon/X_{\alpha\beta}$ and $\sigma_0 := \sigma/X_{\alpha\beta}$ as given and constant.

Table 3
Summary of spatial stretch-based atomistic energies and interaction forces.

Atomistic pairs	Atomistic triplets	Atomistic tetrads
Spatial internal energy densities		
per unit *material* pair length	per unit *material* triplet area	per unit *material* tetrad volume
$W_0^{\alpha\beta}(\lambda_{\alpha\beta})$	$W_0^{\alpha\beta\gamma}(\lambda_{\alpha\beta\gamma})$	$W_0^{\alpha\beta\gamma\delta}(\lambda_{\alpha\beta\gamma\delta})$
Spatial internal energy contributions		
E^{pair}	E^{triplet}	E^{tetrad}
$\frac{1}{2!}\sum_{\alpha,\beta} W_0^{\alpha\beta}(\lambda_{\alpha\beta})\, X_{\alpha\beta}$	$\frac{1}{3!}\sum_{\alpha,\beta,\gamma} W_0^{\alpha\beta\gamma}(\lambda_{\alpha\beta\gamma})\, X_{\alpha\beta\gamma}$	$\frac{1}{4!}\sum_{\alpha,\beta,\gamma,\delta} W_0^{\alpha\beta\gamma\delta}(\lambda_{\alpha\beta\gamma\delta})\, X_{\alpha\beta\gamma\delta}$
Stretch-based spatial interaction forces		
$\mathbb{k}_\alpha^{\text{pair}} = \frac{1}{1!}\sum_\beta \mathbb{k}_{\alpha\beta}^{\text{pair}}$	$\mathbb{k}_\alpha^{\text{triplet}} = \frac{1}{2!}\sum_{\beta,\gamma} \mathbb{k}_{\alpha\beta\gamma}^{\text{triplet}}$	$\mathbb{k}_\alpha^{\text{tetrad}} = \frac{1}{3!}\sum_{\beta,\gamma,\delta} \mathbb{k}_{\alpha\beta\gamma\delta}^{\text{tetrad}}$
$\mathbb{k}_{\alpha\beta}^{\text{pair}} := -\frac{\partial W_0^{\alpha\beta}}{\partial \mathbb{x}_\alpha} X_{\alpha\beta}$	$\mathbb{k}_{\alpha\beta}^{\text{pair}} := -\frac{\partial W_0^{\alpha\beta}}{\partial \mathbb{x}_\alpha} X_{\alpha\beta}$	$\mathbb{k}_{\alpha\beta}^{\text{pair}} := -\frac{\partial W_0^{\alpha\beta}}{\partial \mathbb{x}_\alpha} X_{\alpha\beta}$
Stretch-based spatial interaction force generators		
$\mathbb{g}_{\alpha\beta}^{\text{pair}} := \frac{\partial W_0^{\alpha\beta}}{\partial \mathbb{x}_{\alpha\beta}} X_{\alpha\beta}$	$\mathbb{g}_{\alpha\beta\gamma}^{\text{triplet}} := \frac{\partial W_0^{\alpha\beta\gamma}}{\partial \mathbb{x}_{\alpha\beta\gamma}} X_{\alpha\beta\gamma}$	$\mathbb{g}_{\alpha\beta\gamma\delta}^{\text{tetrad}} := \frac{\partial W_0^{\alpha\beta\gamma\delta}}{\partial \mathbb{x}_{\alpha\beta\gamma\delta}} X_{\alpha\beta\gamma\delta}$
$\mathbb{k}_{\alpha\beta}^{\text{pair}} = \mathbb{g}_{\alpha\beta}^{\text{pair}}$	$\mathbb{k}_{\alpha\beta\gamma}^{\text{triplet}} = \mathbb{x}_{\beta\gamma} \times \mathbb{g}_{\alpha\beta\gamma}^{\text{triplet}}$	$\mathbb{k}_{\alpha\beta\gamma\delta}^{\text{tetrad}} = \mathbb{x}_{\beta\gamma\delta}\, \mathbb{g}_{\alpha\beta\gamma\delta}^{\text{tetrad}}$
Signed magnitudes of stretch-based spatial interaction force generators		
$k_{\alpha\beta}^{\text{pair}} := \frac{\partial W_0^{\alpha\beta}}{\partial \lambda_{\alpha\beta}}$	$k_{\alpha\beta\gamma}^{\text{triplet}} := \frac{\partial W_0^{\alpha\beta\gamma}}{\partial \lambda_{\alpha\beta\gamma}}$	$k_{\alpha\beta\gamma\delta}^{\text{tetrad}} := \frac{\partial W_0^{\alpha\beta\gamma\delta}}{\partial \lambda_{\alpha\beta\gamma\delta}}$
Dimensional reduction to a *membrane* (in-plane stiffness only) with the atomistic positions constrained to a two-dimensional manifold, the tetrad potential is simply neglected by setting $E^{\text{tetrad}} \equiv 0$		
For a dimensional reduction to a *bar* (in-line stiffness only) with the atomistic positions constrained to a one-dimensional manifold, the triplet and tetrad potentials are simply neglected by setting $E^{\text{triplet}} = E^{\text{tetrad}} \equiv 0$		

Remark. For a dimensional reduction to a *membrane* (in-plane stiffness only) with the atomistic positions constrained to a two-dimensional manifold, see Fig. 11, the tetrad potential is simply neglected by setting $E^{\text{tetrad}} \equiv 0$. □

Remark. For a dimensional reduction to a *bar* (in-line stiffness only) with the atomistic positions constrained to a one-dimensional manifold, see Fig. 12, the triplet and tetrad potentials are neglected by setting $E^{\text{triplet}} = E^{\text{tetrad}} \equiv 0$. □

4.2. Spatial stretch-based interaction forces

Due to the stretch-based parametrisation of the pair, triplet and tetrad potentials, the corresponding (resultant) spatial atomistic forces expand as sums of corresponding spatial *interaction forces* as

$$\mathbb{k}_\alpha^{\text{pair}} = \frac{1}{1!}\sum_\beta \mathbb{k}_{\alpha\beta}^{\text{pair}}, \qquad \mathbb{k}_\alpha^{\text{triplet}} = \frac{1}{2!}\sum_{\beta,\gamma} \mathbb{k}_{\alpha\beta\gamma}^{\text{triplet}} \quad \text{and} \quad \mathbb{k}_\alpha^{\text{tetrad}} = \frac{1}{3!}\sum_{\beta,\gamma,\delta} \mathbb{k}_{\alpha\beta\gamma\delta}^{\text{tetrad}}, \tag{34}$$

whereby the stretch-based spatial pair, triplet and tetrad interaction forces follow obviously as

$$\mathbb{k}_{\alpha\beta}^{\text{pair}} := -\frac{\partial W_0^{\alpha\beta}}{\partial \mathbb{x}_\alpha} X_{\alpha\beta}, \qquad \mathbb{k}_{\alpha\beta\gamma}^{\text{triplet}} := -\frac{\partial W_0^{\alpha\beta\gamma}}{\partial \mathbb{x}_\alpha} X_{\alpha\beta\gamma} \quad \text{and} \quad \mathbb{k}_{\alpha\beta\gamma\delta}^{\text{tetrad}} := -\frac{\partial W_0^{\alpha\beta\gamma\delta}}{\partial \mathbb{x}_\alpha} X_{\alpha\beta\gamma\delta}. \tag{35}$$

After some straightforward manipulations, these interaction forces can be stated in terms of the stretch-based spatial pair, triplet and tetrad *interaction force generators* $\mathbb{g}_{\alpha\beta}^{\text{pair}}$, $\mathbb{g}_{\alpha\beta\gamma}^{\text{triplet}}$ and $\mathbb{g}_{\alpha\beta\gamma\delta}^{\text{tetrad}}$ as

$$\mathbb{k}_{\alpha\beta}^{\text{pair}} = \mathbb{g}_{\alpha\beta}^{\text{pair}}, \qquad \mathbb{k}_{\alpha\beta\gamma}^{\text{triplet}} = \mathbb{x}_{\beta\gamma} \times \mathbb{g}_{\alpha\beta\gamma}^{\text{triplet}} \quad \text{and} \quad \mathbb{k}_{\alpha\beta\gamma\delta}^{\text{tetrad}} = \mathbb{x}_{\beta\gamma\delta}\, \mathbb{g}_{\alpha\beta\gamma\delta}^{\text{tetrad}}. \tag{36}$$

In the above, we defined the stretch-based spatial pair, triplet and tetrad interaction force generators as the pair-, triplet- and tetrad-wise sensitivities of the stretch-based pair, triplet and tetrad potentials, respectively. Thus

$$\begin{aligned} \mathbb{g}_{\alpha\beta}^{\text{pair}} &:= \frac{\partial W_0^{\alpha\beta}}{\partial \mathbb{x}_{\alpha\beta}} X_{\alpha\beta} \\ &=: k_{\alpha\beta}^{\text{pair}}\ \mathbb{n}_{\alpha\beta} \end{aligned}, \qquad \begin{aligned} \mathbb{g}_{\alpha\beta\gamma}^{\text{triplet}} &:= \frac{\partial W_0^{\alpha\beta\gamma}}{\partial \mathbb{x}_{\alpha\beta\gamma}} X_{\alpha\beta\gamma} \\ &=: k_{\alpha\beta\gamma}^{\text{triplet}}\ \mathbb{n}_{\alpha\beta\gamma} \end{aligned} \quad \text{and} \quad \begin{aligned} \mathbb{g}_{\alpha\beta\gamma\delta}^{\text{tetrad}} &:= \frac{\partial W_0^{\alpha\beta\gamma\delta}}{\partial \mathbb{x}_{\alpha\beta\gamma\delta}} X_{\alpha\beta\gamma\delta} \\ &=: \mathsf{k}_{\alpha\beta\gamma\delta}^{\text{tetrad}}\ \mathsf{n}_{\alpha\beta\gamma\delta} \end{aligned}. \tag{37}$$

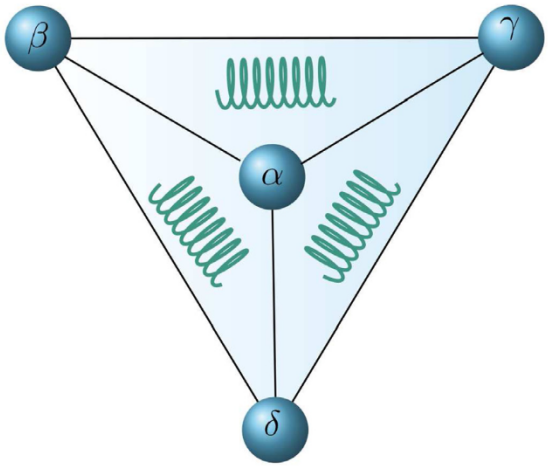


Fig. 11. Atomistic membrane with in-plane stiffness only.

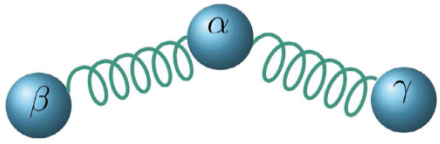


Fig. 12. Atomistic bar with in-line stiffness only.

Therein, we also introduced the *signed magnitudes* of the stretch-based spatial pair, triplet and tetrad interaction force generators, respectively, as[6]

$$k^{\text{pair}}_{\alpha\beta} := \frac{\partial W_0^{\alpha\beta}}{\partial \lambda_{\alpha\beta}}, \qquad k^{\text{triplet}}_{\alpha\beta\gamma} := \frac{\partial W_0^{\alpha\beta\gamma}}{\partial \lambda_{\alpha\beta\gamma}} \quad \text{and} \quad \mathsf{k}^{\text{tetrad}}_{\alpha\beta\gamma\delta} := \frac{\partial W_0^{\alpha\beta\gamma\delta}}{\partial \lambda_{\alpha\beta\gamma\delta}}. \tag{38}$$

Since their signed magnitudes are fully symmetric, the stretch-based spatial pair, triplet and tetrad interaction force generators display the full skew-symmetries

$$\mathbb{g}^{\text{pair}}_{\alpha\beta} \equiv \mathbb{g}^{\text{pair}}_{[\alpha\beta]}, \qquad \mathbb{g}^{\text{triplet}}_{\alpha\beta\gamma} \equiv \mathbb{g}^{\text{triplet}}_{[\alpha\beta\gamma]} \quad \text{and} \quad \mathsf{g}^{\text{tetrad}}_{\alpha\beta\gamma\delta} \equiv \mathsf{g}^{\text{tetrad}}_{[\alpha\beta\gamma\delta]}, \tag{39}$$

moreover, even permutations of index triplets render

$$3\,\mathbb{g}^{\text{triplet}}_{\alpha\beta\gamma} \equiv \mathbb{g}^{\text{triplet}}_{\langle\alpha\beta\gamma\rangle} \quad \text{and} \quad 3\,\mathsf{g}^{\text{tetrad}}_{\alpha\beta\gamma\delta} \equiv \mathsf{g}^{\text{tetrad}}_{\alpha\langle\beta\gamma\delta\rangle}. \tag{40}$$

Remark. The stretch-based spatial pair interaction force $\mathbb{k}^{\text{pair}}_{\alpha\beta}$ is the force exerted on atom α due to its interaction with atom β. It is oriented along the line connecting the pair α, β. The stretch-based spatial triplet interaction force $\mathbb{k}^{\text{triplet}}_{\alpha\beta\gamma}$ is the force exerted on atom α due to its interaction with atoms β and γ. It lies in the plane spanned by the triplet α, β, γ and is oriented perpendicular to the line connecting the pair β, γ. The stretch-based spatial tetrad interaction force $\mathbb{k}^{\text{tetrad}}_{\alpha\beta\gamma\delta}$ is the force exerted on atom α due to its interaction with atoms β, γ and δ. It is oriented perpendicular to the plane spanned by the triplet β, γ, δ. □

Remark. It is interesting to note that due to the equality of the spatial pair, triplet and tetrad stretches associated with all the respective atoms of a particular atomistic pair, triplet and tetrad, the corresponding interaction forces satisfy, respectively, pair-, triplet- and tetrad-wise *actio est reactio* conditions, see Fig. 13. Thereby, the pair-wise *actio est reactio* condition expands as

$$\mathbb{k}^{\text{pair}}_{(\alpha\beta)} \quad \rightarrow \quad \mathbb{n}_{\alpha\beta} + \mathbb{n}_{\beta\alpha} \equiv \mathbb{0}. \tag{41}$$

Likewise, the triplet-wise *actio est reactio* condition reads concisely as

$$\mathbb{k}^{\text{triplet}}_{\langle\alpha\beta\gamma\rangle} \quad \rightarrow \quad \mathbb{x}_{\beta\gamma} + \mathbb{x}_{\gamma\alpha} + \mathbb{x}_{\alpha\beta} \equiv \mathbb{0}. \tag{42}$$

[6] Observe the striking formal similarity of $k^{\text{pair}}_{\alpha\beta}$, $k^{\text{triplet}}_{\alpha\beta\gamma}$ and $\mathsf{k}^{\text{tetrad}}_{\alpha\beta\gamma\delta}$ with the definition of the Piola stress $\boldsymbol{P} := \partial_{\boldsymbol{F}} W_0$ in continuum hyperelasticity, whereby W_0 denotes the energy storage density per unit volume in the material configuration.

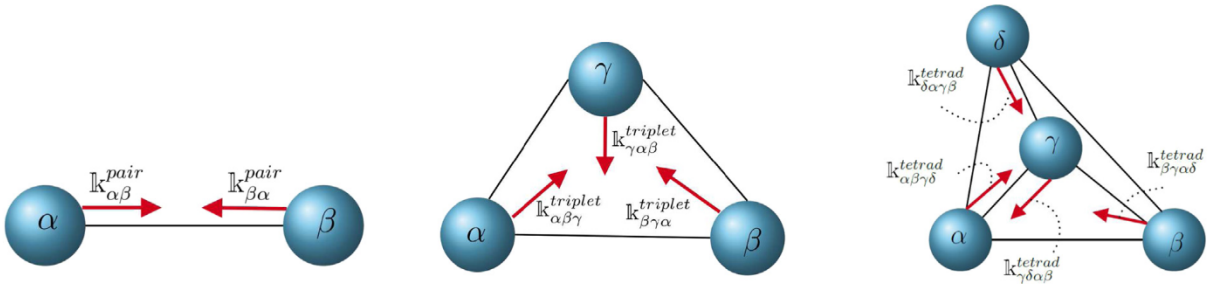


Fig. 13. Actio est reactio in atomistic pairs, triplets and tetrads.

Recall that $\mathbb{x}_{\beta\gamma} + \mathbb{x}_{\gamma\alpha} + \mathbb{x}_{\alpha\beta} = \mathbb{0}$ reflects the *line theorem* equating the integral of the vector-valued line element along a closed circuit to zero.

Finally, the tetrad-wise *actio est reactio* condition follows compactly as

$$\mathbb{k}^{\text{tetrad}}_{\alpha\langle\beta\gamma\delta\rangle} + \mathbb{k}^{\text{tetrad}}_{\beta\langle\gamma\alpha\delta\rangle} + \mathbb{k}^{\text{tetrad}}_{\gamma\langle\delta\alpha\beta\rangle} + \mathbb{k}^{\text{tetrad}}_{\delta\langle\alpha\gamma\beta\rangle} \quad \rightarrow \quad \mathbb{x}_{\beta\gamma\delta} + \mathbb{x}_{\gamma\alpha\delta} + \mathbb{x}_{\delta\alpha\beta} + \mathbb{x}_{\alpha\gamma\beta} \equiv \mathbb{0}. \tag{43}$$

Recall that $\mathbb{x}_{\beta\gamma\delta} + \mathbb{x}_{\gamma\alpha\delta} + \mathbb{x}_{\delta\alpha\beta} + \mathbb{x}_{\alpha\gamma\beta} = \mathbb{0}$ reflects the *area theorem* equating the integral of the vector-valued area element over a closed surface to zero. □

Remark. As an alternative route to derive the stretch-based spatial atomistic forces and the corresponding equilibrium condition, the *principle of spatial atomistic virtual work* equates the spatial internal and external atomistic virtual work for all admissible spatial virtual atomistic displacements as

$$P^{\text{int}}_{\delta} = P^{\text{ext}}_{\delta} \quad \forall\ \mathrm{D}_{\delta}\mathbb{x}_{\alpha} \quad \text{with} \quad P^{\text{ext}}_{\delta} := \sum_{\alpha} \mathbb{k}^{\text{ext}}_{\alpha} \cdot \mathrm{D}_{\delta}\mathbb{x}_{\alpha}. \tag{44}$$

The spatial internal atomistic virtual work decomposes into pair, triplet and tetrad contributions as follows

$$P^{\text{int}}_{\delta} := P^{\text{pair}}_{\delta} + P^{\text{triplet}}_{\delta} + P^{\text{tetrad}}_{\delta}. \tag{45}$$

The pair-, triplet- and tetrad-wise contributions to the spatial internal atomistic virtual work expand in terms of the stretch-based spatial pair, triplet and tetrad interaction force generators $\mathbb{g}^{\text{pair}}_{\alpha\beta}$, $\mathbb{g}^{\text{triplet}}_{\alpha\beta\gamma}$ and $\mathbb{g}^{\text{tetrad}}_{\alpha\beta\gamma\delta}$ as

$$\begin{aligned}
P^{\text{pair}}_{\delta} &:= \frac{1}{2!} \sum_{\alpha,\beta} \mathbb{g}^{\text{pair}}_{\alpha\beta} \cdot \mathrm{D}_{\delta}\mathbb{x}_{\alpha\beta} \\
P^{\text{triplet}}_{\delta} &:= \frac{1}{3!} \sum_{\alpha,\beta,\gamma} \mathbb{g}^{\text{triplet}}_{\alpha\beta\gamma} \cdot \mathrm{D}_{\delta}\mathbb{x}_{\alpha\beta\gamma} \\
P^{\text{tetrad}}_{\delta} &:= \frac{1}{4!} \sum_{\alpha,\beta,\gamma,\delta} \mathbb{g}^{\text{tetrad}}_{\alpha\beta\gamma\delta} \cdot \mathrm{D}_{\delta}\mathbb{x}_{\alpha\beta\gamma\delta}.
\end{aligned} \tag{46}$$

Tedious but straightforward manipulations (detailed in Appendix A.1) that correspond to partial integration[7] (and the application of the Gauss theorem) in the continuum setting yield

$$\begin{aligned}
P^{\text{pair}}_{\delta} &= -\sum_{\alpha} \mathbb{k}^{\text{pair}}_{\alpha} \cdot \mathrm{D}_{\delta}\mathbb{x}_{\alpha} := -\frac{1}{1!} \sum_{\alpha,\beta} \mathbb{k}^{\text{pair}}_{\alpha\beta} \cdot \mathrm{D}_{\delta}\mathbb{x}_{\alpha}, \\
P^{\text{triplet}}_{\delta} &= -\sum_{\alpha} \mathbb{k}^{\text{triplet}}_{\alpha} \cdot \mathrm{D}_{\delta}\mathbb{x}_{\alpha} := -\frac{1}{2!} \sum_{\alpha,\beta,\gamma} \mathbb{k}^{\text{triplet}}_{\alpha\beta\gamma} \cdot \mathrm{D}_{\delta}\mathbb{x}_{\alpha}, \\
P^{\text{tetrad}}_{\delta} &= -\sum_{\alpha} \mathbb{k}^{\text{tetrad}}_{\alpha} \cdot \mathrm{D}_{\delta}\mathbb{x}_{\alpha} := -\frac{1}{3!} \sum_{\alpha,\beta,\gamma,\delta} \mathbb{k}^{\text{tetrad}}_{\alpha\beta\gamma\delta} \cdot \mathrm{D}_{\delta}\mathbb{x}_{\alpha}.
\end{aligned} \tag{47}$$

Thus, the principle of spatial atomistic virtual work is equivalent to the spatial atomistic equilibrium condition $\mathbb{k}^{\text{int}}_{\alpha} + \mathbb{k}^{\text{ext}}_{\alpha} = \mathbb{0}$ with $\mathbb{k}^{\text{int}}_{\alpha} := \mathbb{k}^{\text{pair}}_{\alpha} + \mathbb{k}^{\text{triplet}}_{\alpha} + \mathbb{k}^{\text{tetrad}}_{\alpha}$, whereby the stretch-based spatial pair, triplet and tetrad interaction forces follow in the already established fashion from the corresponding interaction force generators. For a related discussion regarding the classical mechanics of particle systems see Fried (2010). □

Remark. For a dimensional reduction to a *membrane* (in-plane stiffness only) with the atomistic positions constrained to a two-dimensional manifold, the stretch-based spatial tetrad interaction forces are simply neglected by setting $\mathbb{k}^{\text{tetrad}}_{\alpha\beta\gamma\delta} \equiv \mathbb{0}$. □

[7] $\boldsymbol{P} : \nabla_X \mathrm{D}_{\delta}\boldsymbol{y} = -\mathrm{D}_{\delta}\boldsymbol{y} \cdot \mathrm{Div}\boldsymbol{P} + \mathrm{Div}(\mathrm{D}_{\delta}\boldsymbol{y} \cdot \boldsymbol{P})$.

Remark. For a dimensional reduction to a *bar* (in-line stiffness only) with the atomistic positions constrained to a one-dimensional manifold, the stretch-based spatial triplet and tetrad interaction forces are simply neglected by setting $\Bbbk^{\text{triplet}}_{\alpha\beta\gamma} = \Bbbk^{\text{tetrad}}_{\alpha\beta\gamma\delta} \equiv \mathbb{0}$. □

4.3. Material stretch-based parametrisation

For the material setting we alternatively consider the atomistic pair, triplet and tetrad contributions – E^{pair}, E^{triplet} and E^{tetrad} – to the internal potential energy E^{int} in terms of their densities – $W_t^{\alpha\beta}$, $W_t^{\alpha\beta\gamma}$ and $W_t^{\alpha\beta\gamma\delta}$ – per unit *spatial* pair length $x_{\alpha\beta}$, triplet area $x_{\alpha\beta\gamma}$ and tetrad volume $x_{\alpha\beta\gamma\delta}$, respectively. We parameterise these energy densities in terms of material pair, triplet and tetrad stretches $\Lambda_{\alpha\beta}$, $\Lambda_{\alpha\beta\gamma}$ and $\Lambda_{\alpha\beta\gamma\delta}$, respectively. This yields

$$E^{\text{pair}} := \frac{1}{2!}\sum_{\alpha,\beta} W_t^{\alpha\beta}(\Lambda_{\alpha\beta})\, x_{\alpha\beta} \tag{48}$$

$$E^{\text{triplet}} := \frac{1}{3!}\sum_{\alpha,\beta,\gamma} W_t^{\alpha\beta\gamma}(\Lambda_{\alpha\beta\gamma})\, x_{\alpha\beta\gamma} \tag{49}$$

$$E^{\text{tetrad}} := \frac{1}{4!}\sum_{\alpha,\beta,\gamma,\delta} W_t^{\alpha\beta\gamma\delta}(\Lambda_{\alpha\beta\gamma\delta})\, x_{\alpha\beta\gamma\delta}. \tag{50}$$

Observe that the spatial and material potential energy densities are related as

$$W_t^{\alpha\beta} = \Lambda_{\alpha\beta}\, W_0^{\alpha\beta}, \qquad W_t^{\alpha\beta\gamma} = \Lambda_{\alpha\beta\gamma}\, W_0^{\alpha\beta\gamma} \quad \text{and} \quad W_t^{\alpha\beta\gamma\delta} = \Lambda_{\alpha\beta\gamma\delta}\, W_0^{\alpha\beta\gamma\delta}, \tag{51}$$

whereby the material pair, triplet and tetrad stretches $\Lambda_{\alpha\beta}$, $\Lambda_{\alpha\beta\gamma}$ and $\Lambda_{\alpha\beta\gamma\delta}$, respectively, serve to transform material densities into spatial densities.

4.4. Material stretch-based interaction forces

The (resultant) material atomistic forces corresponding to the stretch-based pair, triplet and tetrad potentials can also be expanded as sums of corresponding material interaction forces:

$$\mathbb{K}^{\text{pair}}_{\alpha} = \frac{1}{1!}\sum_{\beta} \mathbb{K}^{\text{pair}}_{\alpha\beta}, \qquad \mathbb{K}^{\text{triplet}}_{\alpha} = \frac{1}{2!}\sum_{\beta,\gamma} \mathbb{K}^{\text{triplet}}_{\alpha\beta\gamma} \quad \text{and} \quad \mathbb{K}^{\text{tetrad}}_{\alpha} = \frac{1}{3!}\sum_{\beta,\gamma,\delta} \mathbb{K}^{\text{tetrad}}_{\alpha\beta\gamma\delta} \tag{52}$$

with the stretch-based material pair, triplet and tetrad interaction forces given by

$$\mathbb{K}^{\text{pair}}_{\alpha\beta} := -\frac{\partial W_t^{\alpha\beta}}{\partial \mathbb{X}_{\alpha}}\, x_{\alpha\beta}, \qquad \mathbb{K}^{\text{triplet}}_{\alpha\beta\gamma} := -\frac{\partial W_t^{\alpha\beta\gamma}}{\partial \mathbb{X}_{\alpha}}\, x_{\alpha\beta\gamma} \quad \text{and} \quad \mathbb{K}^{\text{tetrad}}_{\alpha\beta\gamma\delta} := -\frac{\partial W_t^{\alpha\beta\gamma\delta}}{\partial \mathbb{X}_{\alpha}}\, x_{\alpha\beta\gamma\delta}. \tag{53}$$

In analogy to the spatial interaction forces, the material interaction forces can be expressed in terms of stretch-based material pair, triplet and tetrad interaction force generators $\mathbb{G}^{\text{pair}}_{\alpha\beta}$, $\mathbb{G}^{\text{triplet}}_{\alpha\beta\gamma}$ and $\mathsf{G}^{\text{tetrad}}_{\alpha\beta\gamma\delta}$ as

$$\mathbb{K}^{\text{pair}}_{\alpha\beta} = \mathbb{G}^{\text{pair}}_{\alpha\beta}, \qquad \mathbb{K}^{\text{triplet}}_{\alpha\beta\gamma} = \mathbb{X}_{\beta\gamma} \times \mathbb{G}^{\text{triplet}}_{\alpha\beta\gamma} \quad \text{and} \quad \mathbb{K}^{\text{tetrad}}_{\alpha\beta\gamma\delta} = \mathbb{X}_{\beta\gamma\delta}\, \mathsf{G}^{\text{tetrad}}_{\alpha\beta\gamma\delta}. \tag{54}$$

Likewise, in analogy to the spatial interaction force generators, the stretch-based material pair, triplet and tetrad interaction force generators follow as the pair-, triplet- and tetrad-wise sensitivities of the stretch-based pair, triplet and tetrad potentials, respectively. That is

$$\begin{aligned} \mathbb{G}^{\text{pair}}_{\alpha\beta} &:= \frac{\partial W_t^{\alpha\beta}}{\partial \mathbb{X}_{\alpha\beta}}\, x_{\alpha\beta} \\ &=: K^{\text{pair}}_{\alpha\beta}\ \mathbb{N}_{\alpha\beta} \end{aligned}, \qquad \begin{aligned} \mathbb{G}^{\text{triplet}}_{\alpha\beta\gamma} &:= \frac{\partial W_t^{\alpha\beta\gamma}}{\partial \mathbb{X}_{\alpha\beta\gamma}}\, x_{\alpha\beta\gamma} \\ &=: K^{\text{triplet}}_{\alpha\beta\gamma}\ \mathbb{N}_{\alpha\beta\gamma} \end{aligned} \quad \text{and} \quad \begin{aligned} \mathsf{G}^{\text{tetrad}}_{\alpha\beta\gamma\delta} &:= \frac{\partial W_t^{\alpha\beta\gamma\delta}}{\partial \mathsf{X}_{\alpha\beta\gamma\delta}}\, x_{\alpha\beta\gamma\delta} \\ &=: \mathsf{K}^{\text{tetrad}}_{\alpha\beta\gamma\delta}\ \mathsf{N}_{\alpha\beta\gamma\delta} \end{aligned}. \tag{55}$$

Here, the signed magnitudes of the stretch-based material pair, triplet and tetrad interaction force generators follow as

$$K^{\text{pair}}_{\alpha\beta} := \frac{\partial W_t^{\alpha\beta}}{\partial \Lambda_{\alpha\beta}}, \qquad K^{\text{triplet}}_{\alpha\beta\gamma} := \frac{\partial W_t^{\alpha\beta\gamma}}{\partial \Lambda_{\alpha\beta\gamma}} \quad \text{and} \quad \mathsf{K}^{\text{tetrad}}_{\alpha\beta\gamma\delta} := \frac{\partial W_t^{\alpha\beta\gamma\delta}}{\partial \Lambda_{\alpha\beta\gamma\delta}}. \tag{56}$$

It is worth noticing that these force generators can be expanded in terms of their spatial counterparts as

$$K^{\text{pair}}_{\alpha\beta} = W_0^{\alpha\beta} - \lambda_{\alpha\beta}\, k^{\text{pair}}_{\alpha\beta}, \quad K^{\text{triplet}}_{\alpha\beta\gamma} = W_0^{\alpha\beta\gamma} - \lambda_{\alpha\beta\gamma}\, k^{\text{triplet}}_{\alpha\beta\gamma} \quad \text{and} \quad \mathsf{K}^{\text{tetrad}}_{\alpha\beta\gamma\delta} = W_0^{\alpha\beta\gamma\delta} - \lambda_{\alpha\beta\gamma\delta}\, \mathsf{k}^{\text{tetrad}}_{\alpha\beta\gamma\delta}. \tag{57}$$

Thus, the classical energy–momentum format of the Eshelby stress[8] established in the continuum setting is recovered in the expressions of the signed magnitudes of the stretch-based material pair, triplet and tetrad interaction force generators. We consider this formal similarity striking!

[8] $\boldsymbol{\Sigma} := W_0 \boldsymbol{I} - \boldsymbol{F}^t \cdot \boldsymbol{P}$.

Table 4
Summary of material stretch-based atomistic energies and interaction forces.

Atomistic pairs	Atomistic triplets	Atomistic tetrads
Material internal energy densities		
per unit *spatial* pair length	per unit *spatial* triplet area	per unit *spatial* tetrad volume
$W_t^{\alpha\beta}(\Lambda_{\alpha\beta}) = \Lambda_{\alpha\beta}\, W_0^{\alpha\beta}$	$W_t^{\alpha\beta\gamma}(\Lambda_{\alpha\beta\gamma}) = \Lambda_{\alpha\beta\gamma}\, W_0^{\alpha\beta\gamma}$	$W_t^{\alpha\beta\gamma\delta}(\Lambda_{\alpha\beta\gamma\delta}) = \Lambda_{\alpha\beta\gamma\delta}\, W_0^{\alpha\beta\gamma\delta}$
Material internal energy contributions		
E^{pair}	E^{triplet}	E^{tetrad}
$\frac{1}{2!}\sum_{\alpha,\beta} W_t^{\alpha\beta}(\Lambda_{\alpha\beta})\, x_{\alpha\beta}$	$\frac{1}{3!}\sum_{\alpha,\beta,\gamma} W_t^{\alpha\beta\gamma}(\Lambda_{\alpha\beta\gamma})\, x_{\alpha\beta\gamma}$	$\frac{1}{4!}\sum_{\alpha,\beta,\gamma,\delta} W_t^{\alpha\beta\gamma\delta}(\Lambda_{\alpha\beta\gamma\delta})\, x_{\alpha\beta\gamma\delta}$
Stretch-based material interaction forces		
$\mathbb{K}_\alpha^{\text{pair}} = \frac{1}{1!}\sum_{\beta} \mathbb{K}_{\alpha\beta}^{\text{pair}}$	$\mathbb{K}_\alpha^{\text{triplet}} = \frac{1}{2!}\sum_{\beta,\gamma} \mathbb{K}_{\alpha\beta\gamma}^{\text{triplet}}$	$\mathbb{K}_\alpha^{\text{tetrad}} = \frac{1}{3!}\sum_{\beta,\gamma,\delta} \mathbb{K}_{\alpha\beta\gamma\delta}^{\text{tetrad}}$
$\mathbb{K}_{\alpha\beta}^{\text{pair}} := -\frac{\partial W_t^{\alpha\beta}}{\partial \mathbb{X}_\alpha}\, x_{\alpha\beta}$	$\mathbb{K}_{\alpha\beta}^{\text{pair}} := -\frac{\partial W_t^{\alpha\beta}}{\partial \mathbb{X}_\alpha}\, x_{\alpha\beta}$	$\mathbb{K}_{\alpha\beta}^{\text{pair}} := -\frac{\partial W_t^{\alpha\beta}}{\partial \mathbb{X}_\alpha}\, x_{\alpha\beta}$
Stretch-based material interaction force generators		
$\mathbb{G}_{\alpha\beta}^{\text{pair}} := \frac{\partial W_t^{\alpha\beta}}{\partial \mathbb{X}_{\alpha\beta}}\, x_{\alpha\beta}$	$\mathbb{G}_{\alpha\beta\gamma}^{\text{triplet}} := \frac{\partial W_t^{\alpha\beta\gamma}}{\partial \mathbb{X}_{\alpha\beta\gamma}}\, x_{\alpha\beta\gamma}$	$\mathsf{G}_{\alpha\beta\gamma\delta}^{\text{tetrad}} := \frac{\partial W_t^{\alpha\beta\gamma\delta}}{\partial \mathsf{X}_{\alpha\beta\gamma\delta}}\, x_{\alpha\beta\gamma\delta}$
$\mathbb{K}_{\alpha\beta}^{\text{pair}} = \mathbb{G}_{\alpha\beta}^{\text{pair}}$	$\mathbb{K}_{\alpha\beta\gamma}^{\text{triplet}} = \mathbb{X}_{\beta\gamma} \times \mathbb{G}_{\alpha\beta\gamma}^{\text{triplet}}$	$\mathbb{K}_{\alpha\beta\gamma\delta}^{\text{tetrad}} = \mathbb{X}_{\beta\gamma\delta}\, \mathsf{G}_{\alpha\beta\gamma\delta}^{\text{tetrad}}$
Signed magnitudes of stretch-based material interaction force generators		
$K_{\alpha\beta}^{\text{pair}} := \frac{\partial W_t^{\alpha\beta}}{\partial \Lambda_{\alpha\beta}}$	$K_{\alpha\beta\gamma}^{\text{triplet}} := \frac{\partial W_t^{\alpha\beta\gamma}}{\partial \Lambda_{\alpha\beta\gamma}}$	$\mathsf{K}_{\alpha\beta\gamma\delta}^{\text{tetrad}} := \frac{\partial W_t^{\alpha\beta\gamma\delta}}{\partial \Lambda_{\alpha\beta\gamma\delta}}$
Relationship between signed magnitudes of spatial and material interaction force generators		
$K_{\alpha\beta}^{\text{pair}} = W_0^{\alpha\beta} - \lambda_{\alpha\beta}\, k_{\alpha\beta}^{\text{pair}}$	$K_{\alpha\beta\gamma}^{\text{triplet}} = W_0^{\alpha\beta\gamma} - \lambda_{\alpha\beta\gamma}\, k_{\alpha\beta\gamma}^{\text{triplet}}$	$\mathsf{K}_{\alpha\beta\gamma\delta}^{\text{tetrad}} = W_0^{\alpha\beta\gamma\delta} - \lambda_{\alpha\beta\gamma\delta}\, \mathsf{k}_{\alpha\beta\gamma\delta}^{\text{tetrad}}$

5. Twist-based energies

5.1. Spatial twist-based parametrisation

Here, we consider an alternative expansion of the atomistic triplet and tetrad contributions – E^{triplet} and E^{tetrad} – to the internal potential energy E^{int} in terms of their densities – $w_0^{\alpha\beta\gamma}$ and $w_0^{\alpha\beta\gamma\delta}$ – per unit *material* angle $\boldsymbol{\Phi}_{\alpha\beta\gamma}$ and dihedral angle $\boldsymbol{\Phi}_{\alpha\beta\gamma\delta}$, respectively. We parameterise these energy densities in terms of spatial triplet and tetrad twists $\omega_{\alpha\beta\gamma}$ and $\omega_{\alpha\beta\gamma\delta}$, respectively, thus[9]

$$E^{\text{triplet}} := \frac{1}{3!}\sum_{\alpha,\beta,\gamma} \Big[\, w_0^{\alpha\beta\gamma}(\omega_{\alpha\beta\gamma})\,\boldsymbol{\Phi}_{\alpha\beta\gamma} + w_0^{\beta\gamma\alpha}(\omega_{\beta\gamma\alpha})\,\boldsymbol{\Phi}_{\beta\gamma\alpha} + w_0^{\gamma\alpha\beta}(\omega_{\gamma\alpha\beta})\,\boldsymbol{\Phi}_{\gamma\alpha\beta} \,\Big] \tag{58}$$

$$\begin{aligned} E^{\text{tetrad}} := \frac{1}{4!}\sum_{\alpha,\beta,\gamma,\delta} \Big[\, & w_0^{\alpha\delta\beta\gamma}(\omega_{\alpha\delta\beta\gamma})\,\boldsymbol{\Phi}_{\alpha\delta\beta\gamma} + w_0^{\alpha\beta\gamma\delta}(\omega_{\alpha\beta\gamma\delta})\,\boldsymbol{\Phi}_{\alpha\beta\gamma\delta} + w_0^{\alpha\gamma\delta\beta}(\omega_{\alpha\gamma\delta\beta})\,\boldsymbol{\Phi}_{\alpha\gamma\delta\beta} \\ & + w_0^{\beta\alpha\delta\gamma}(\omega_{\beta\alpha\delta\gamma})\,\boldsymbol{\Phi}_{\beta\alpha\delta\gamma} + w_0^{\beta\delta\gamma\alpha}(\omega_{\beta\delta\gamma\alpha})\,\boldsymbol{\Phi}_{\beta\delta\gamma\alpha} + w_0^{\beta\gamma\alpha\delta}(\omega_{\beta\gamma\alpha\delta})\,\boldsymbol{\Phi}_{\beta\gamma\alpha\delta} \\ & + w_0^{\gamma\beta\delta\alpha}(\omega_{\gamma\beta\delta\alpha})\,\boldsymbol{\Phi}_{\gamma\beta\delta\alpha} + w_0^{\gamma\delta\alpha\beta}(\omega_{\gamma\delta\alpha\beta})\,\boldsymbol{\Phi}_{\gamma\delta\alpha\beta} + w_0^{\gamma\alpha\beta\delta}(\omega_{\gamma\alpha\beta\delta})\,\boldsymbol{\Phi}_{\gamma\alpha\beta\delta} \\ & + w_0^{\delta\gamma\beta\alpha}(\omega_{\delta\gamma\beta\alpha})\,\boldsymbol{\Phi}_{\delta\gamma\beta\alpha} + w_0^{\delta\beta\alpha\gamma}(\omega_{\delta\beta\alpha\gamma})\,\boldsymbol{\Phi}_{\delta\beta\alpha\gamma} + w_0^{\delta\alpha\gamma\beta}(\omega_{\delta\alpha\gamma\beta})\,\boldsymbol{\Phi}_{\delta\alpha\gamma\beta} \,\Big]. \end{aligned}$$

Recall that the spatial triplet and tetrad twists $\omega_{\alpha\beta\gamma}$ and $\omega_{\alpha\beta\gamma\delta}$, respectively, are in general distinct for all atoms belonging to a specific atomistic triplet and tetrad, respectively. As a result their contribution is included in each summand of the above expansions.

Remark. For a dimensional reduction to a *shell* (bending stiffness only) with the atomistic positions constrained to a two-dimensional manifold, see Fig. 14, the tetrad potential E^{tetrad} is reduced by setting $w_0^{\beta\delta\gamma\alpha} = w_0^{\delta\beta\alpha\gamma} = w_0^{\gamma\beta\delta\alpha} = w_0^{\beta\gamma\alpha\delta} = w_0^{\delta\gamma\beta\alpha} = w_0^{\gamma\delta\alpha\beta} \equiv 0$. □

[9] Exploiting the left and right minor symmetries of the tetrad twists $\omega_{\alpha\beta\gamma\delta} = \omega_{(\alpha\beta)(\gamma\delta)}$ allows a reduction in the number of terms from 12 to 6. We prefer, however, to retain all 12 permutations of $\alpha, \beta, \gamma, \delta$ in the derivations for the sake of generality, a choice that eventually reflects in the pre-factor 2 when expressing the corresponding interaction forces.

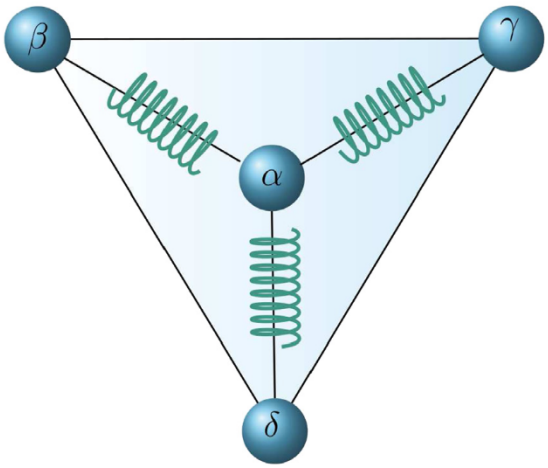


Fig. 14. Atomistic shell with bending stiffness only.

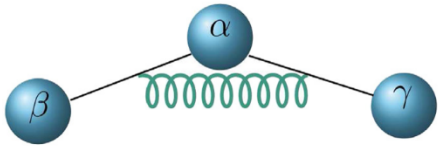


Fig. 15. Atomistic beam with bending stiffness only.

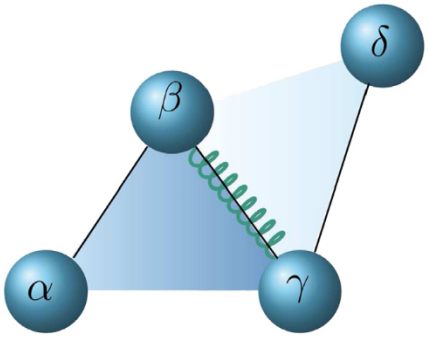


Fig. 16. Atomistic shaft with torsion stiffness only.

Remark. For a dimensional reduction to a *beam* (bending stiffness only) with the atomistic positions constrained to a one-dimensional manifold, see Fig. 15, the triplet potential E^{triplet} is reduced by setting $w_0^{\beta\gamma\alpha} = w_0^{\gamma\alpha\beta} \equiv 0$, while the tetrad potential is simply neglected by setting $E^{\text{tetrad}} \equiv 0$. □

Remark. For a dimensional reduction to a *shaft* (torsion stiffness only) with the atomistic positions constrained to a one-dimensional manifold, see Fig. 16, the tetrad potential E^{tetrad} is reduced by retaining only $w_0^{\beta\gamma\alpha\delta} = w_0^{\gamma\beta\delta\alpha} \neq 0$, while the triplet potential is simply neglected by setting $E^{\text{triplet}} \equiv 0$. □

5.2. Spatial twist-based interaction forces

Due to the twist-based parametrisation of the triplet and tetrad potentials, the corresponding (resultant) spatial atomistic forces expand, as before, as sums of corresponding spatial interaction forces. Thus

$$\mathbb{k}_\alpha^{\text{triplet}} = \frac{1}{2!}\sum_{\beta,\gamma}\mathbb{k}_{\alpha\beta\gamma}^{\text{triplet}} \quad \text{and} \quad \mathbb{k}_\alpha^{\text{tetrad}} = \frac{1}{3!}\sum_{\beta,\gamma,\delta}\mathbb{k}_{\alpha\beta\gamma\delta}^{\text{tetrad}}, \tag{59}$$

where the twist-based spatial triplet and tetrad interaction forces are given by

$$\begin{aligned}
\mathbb{k}^{\text{triplet}}_{\alpha\beta\gamma} &:= -\frac{\partial w_0^{\alpha\beta\gamma}}{\partial \mathbb{x}_\alpha}\,\boldsymbol{\Phi}_{\alpha\beta\gamma} - \frac{\partial w_0^{\beta\gamma\alpha}}{\partial \mathbb{x}_\alpha}\,\boldsymbol{\Phi}_{\beta\gamma\alpha} - \frac{\partial w_0^{\gamma\alpha\beta}}{\partial \mathbb{x}_\alpha}\,\boldsymbol{\Phi}_{\gamma\alpha\beta}\,, \\
\mathbb{k}^{\text{tetrad}}_{\alpha\beta\gamma\delta} &:= -\frac{\partial w_0^{\alpha\delta\beta\gamma}}{\partial \mathbb{x}_\alpha}\,\boldsymbol{\Phi}_{\alpha\delta\beta\gamma} - \frac{\partial w_0^{\alpha\beta\gamma\delta}}{\partial \mathbb{x}_\alpha}\,\boldsymbol{\Phi}_{\alpha\beta\gamma\delta} - \frac{\partial w_0^{\alpha\gamma\delta\beta}}{\partial \mathbb{x}_\alpha}\,\boldsymbol{\Phi}_{\alpha\gamma\delta\beta} \\
&\quad - \frac{\partial w_0^{\beta\alpha\delta\gamma}}{\partial \mathbb{x}_\alpha}\,\boldsymbol{\Phi}_{\beta\alpha\delta\gamma} - \frac{\partial w_0^{\beta\delta\gamma\alpha}}{\partial \mathbb{x}_\alpha}\,\boldsymbol{\Phi}_{\beta\delta\gamma\alpha} - \frac{\partial w_0^{\beta\gamma\alpha\delta}}{\partial \mathbb{x}_\alpha}\,\boldsymbol{\Phi}_{\beta\gamma\alpha\delta} \\
&\quad - \frac{\partial w_0^{\gamma\beta\delta\alpha}}{\partial \mathbb{x}_\alpha}\,\boldsymbol{\Phi}_{\gamma\beta\delta\alpha} - \frac{\partial w_0^{\gamma\delta\alpha\beta}}{\partial \mathbb{x}_\alpha}\,\boldsymbol{\Phi}_{\gamma\delta\alpha\beta} - \frac{\partial w_0^{\gamma\alpha\beta\delta}}{\partial \mathbb{x}_\alpha}\,\boldsymbol{\Phi}_{\gamma\alpha\beta\delta} \\
&\quad - \frac{\partial w_0^{\delta\gamma\beta\alpha}}{\partial \mathbb{x}_\alpha}\,\boldsymbol{\Phi}_{\delta\gamma\beta\alpha} - \frac{\partial w_0^{\delta\beta\alpha\gamma}}{\partial \mathbb{x}_\alpha}\,\boldsymbol{\Phi}_{\delta\beta\alpha\gamma} - \frac{\partial w_0^{\delta\alpha\gamma\beta}}{\partial \mathbb{x}_\alpha}\,\boldsymbol{\Phi}_{\delta\alpha\gamma\beta}.
\end{aligned} \tag{60}$$

After straightforward but lengthy and tedious manipulations (see Appendix B.1), these read in terms of twist-based spatial triplet and tetrad interaction force generators $\mathbb{g}^{\text{triplet}}_{\alpha\beta}$ and $g^{\text{tetrad}}_{\alpha\beta\gamma}$ in compact format as

$$\begin{aligned}
\mathbb{k}^{\text{triplet}}_{\alpha\beta\gamma} &:= 2\big[\mathbb{g}^{\text{triplet}}_{[\alpha\beta]} + \mathbb{g}^{\text{triplet}}_{[\alpha\gamma]}\big], \\
\mathbb{k}^{\text{tetrad}}_{\alpha\beta\gamma\delta} &:= 2\big[\mathbb{x}_{\beta\gamma}\times[\mathbb{g}^{\text{tetrad}}_{[\alpha\beta]\gamma} + \mathbb{g}^{\text{tetrad}}_{[\gamma\alpha]\beta} + \mathbb{g}^{\text{tetrad}}_{[\beta\gamma]\alpha}] \\
&\quad + \mathbb{x}_{\gamma\delta}\times[\mathbb{g}^{\text{tetrad}}_{[\alpha\gamma]\delta} + \mathbb{g}^{\text{tetrad}}_{[\delta\alpha]\gamma} + \mathbb{g}^{\text{tetrad}}_{[\gamma\delta]\alpha}] \\
&\quad + \mathbb{x}_{\delta\beta}\times[\mathbb{g}^{\text{tetrad}}_{[\alpha\delta]\beta} + \mathbb{g}^{\text{tetrad}}_{[\beta\alpha]\delta} + \mathbb{g}^{\text{tetrad}}_{[\delta\beta]\alpha}]\big].
\end{aligned} \tag{61}$$

Note that $\mathbb{g}^{\text{triplet}}_{[\beta\gamma]}$, $\mathbb{g}^{\text{tetrad}}_{[\beta\delta]\gamma}$, $\mathbb{g}^{\text{tetrad}}_{[\gamma\beta]\delta}$ and $\mathbb{g}^{\text{tetrad}}_{[\delta\gamma]\beta}$ (i.e. no index α) do not contribute to the triplet- and tetrad-wise spatial interaction forces $\mathbb{k}^{\text{triplet}}_{\alpha\beta\gamma}$ and $\mathbb{k}^{\text{tetrad}}_{\alpha\beta\gamma\delta}$.

In the above, we defined the twist-based spatial triplet and tetrad interaction force generators as pair- and triplet-wise sensitivities of the twist-based triplet and tetrad potentials. We list the complete sets in Appendix B.2, the following are but two examples

$$\begin{aligned}
\mathbb{g}^{\text{triplet}}_{\alpha\beta} &:= \frac{\partial w_0^{\alpha\beta\gamma}}{\partial \mathbb{x}_{\alpha\beta}}\,\boldsymbol{\Phi}_{\alpha\beta\gamma} \qquad \text{and} \qquad \mathbb{g}^{\text{tetrad}}_{\alpha\beta\gamma} := \frac{\partial w_0^{\alpha\beta\gamma\delta}}{\partial \mathbb{x}_{\alpha\beta\gamma}}\,\boldsymbol{\Phi}_{\alpha\beta\gamma\delta}\,. \\
&=: k^{\text{triplet}}_{\alpha\beta\gamma}\ \mathbb{p}^{\perp}_{\alpha\beta}\cdot\mathbb{m}_{\alpha\gamma} \qquad\qquad =: k^{\text{tetrad}}_{\alpha\beta\gamma\delta}\ \mathbb{p}^{\perp}_{\alpha\beta\gamma}\cdot\mathbb{m}_{\alpha\beta\delta}
\end{aligned} \tag{62}$$

Here we introduced the signed "magnitudes" of the twist-based spatial triplet and tetrad interaction force generators. We list the complete set in Appendix B.2, and only provide the following two examples

$$k^{\text{triplet}}_{\alpha\beta\gamma} := \frac{\partial w_0^{\alpha\beta\gamma}}{\partial \omega_{\alpha\beta\gamma}} \quad \text{and} \quad k^{\text{tetrad}}_{\alpha\beta\gamma\delta} := \frac{\partial w_0^{\alpha\beta\gamma\delta}}{\partial \omega_{\alpha\beta\gamma\delta}}. \tag{63}$$

Since their signed magnitudes display left and right minor symmetries, the twist-based spatial tetrad interaction force generators display left skew-symmetries, for example

$$\mathbb{g}^{\text{tetrad}}_{\alpha\beta\gamma} \equiv \mathbb{g}^{\text{tetrad}}_{[\alpha\beta]\gamma}. \tag{64}$$

Remark. The twist-based spatial triplet interaction force $\mathbb{k}^{\text{triplet}}_{\alpha\beta\gamma}$ is the force exerted on atom α due to its interaction with atoms β and γ. It lies in the plane spanned by the triplet α,β,γ with $\mathbb{g}^{\text{triplet}}_{[\alpha\beta]}$ and $\mathbb{g}^{\text{triplet}}_{[\alpha\gamma]}$ oriented perpendicular to the lines connecting the pairs α,β and α,γ, respectively. The twist-based spatial tetrad interaction force $\mathbb{k}^{\text{tetrad}}_{\alpha\beta\gamma\delta}$ is the force exerted on atom α due to its interaction with atoms β, γ and δ. It is assembled from contributions oriented perpendicular to the three planes spanned by the triplets α,β,γ and α,γ,δ as well as α,δ,β (since, for example, $\mathbb{x}_{\beta\gamma}$ and $\mathbb{g}^{\text{tetrad}}_{\langle\alpha\beta\gamma\rangle}$ lie in the plane spanned by the triplet α,β,γ, etc.). □

Remark. Observe that the spatial twist-based triplet and tetrad interaction forces also satisfy, respectively, triplet- and tetrad-wise *actio est reactio* conditions. Thereby, the triplet-wise *actio est reactio* condition reads as

$$\mathbb{k}^{\text{triplet}}_{\langle\alpha\beta\gamma\rangle} \quad\to\quad \mathbb{g}^{\text{triplet}}_{[\alpha\beta]} + \mathbb{g}^{\text{triplet}}_{[\alpha\gamma]} + \mathbb{g}^{\text{triplet}}_{[\beta\gamma]} + \mathbb{g}^{\text{triplet}}_{[\beta\alpha]} + \mathbb{g}^{\text{triplet}}_{[\gamma\alpha]} + \mathbb{g}^{\text{triplet}}_{[\gamma\beta]} \equiv \mathbb{0}. \tag{65}$$

Likewise, the tetrad-wise *actio est reactio* condition (see Appendix B.3) follows as

$$\begin{aligned}
\mathbb{k}^{\text{tetrad}}_{\alpha\langle\beta\gamma\delta\rangle} + \mathbb{k}^{\text{tetrad}}_{\beta\langle\gamma\alpha\delta\rangle} + \mathbb{k}^{\text{tetrad}}_{\gamma\langle\delta\alpha\beta\rangle} + \mathbb{k}^{\text{tetrad}}_{\delta\langle\alpha\gamma\beta\rangle} \quad\to\quad & [\mathbb{x}_{\beta\gamma} + \mathbb{x}_{\gamma\delta} + \mathbb{x}_{\delta\beta}]\times\mathbb{g}^{\text{tetrad}}_{\langle\beta\gamma\delta\rangle} \\
& + [\mathbb{x}_{\gamma\alpha} + \mathbb{x}_{\alpha\delta} + \mathbb{x}_{\delta\gamma}]\times\mathbb{g}^{\text{tetrad}}_{\langle\gamma\alpha\delta\rangle} \\
& + [\mathbb{x}_{\delta\alpha} + \mathbb{x}_{\alpha\beta} + \mathbb{x}_{\beta\delta}]\times\mathbb{g}^{\text{tetrad}}_{\langle\delta\alpha\beta\rangle} \\
& + [\mathbb{x}_{\alpha\gamma} + \mathbb{x}_{\gamma\beta} + \mathbb{x}_{\beta\alpha}]\times\mathbb{g}^{\text{tetrad}}_{\langle\alpha\gamma\beta\rangle} \equiv \mathbb{0}.
\end{aligned} \tag{66}$$

Recall that, e.g., $\mathbb{x}_{\beta\gamma} + \mathbb{x}_{\gamma\delta} + \mathbb{x}_{\delta\beta} = \mathbb{0}$ reflects the line theorem equating the integral of the vector-valued line element along a closed circuit to zero. □

Remark. The triplet-wise contribution to the spatial internal atomistic virtual work expands in terms of the twist-based spatial triplet interaction force generators $\mathbb{g}^{\text{triplet}}_{\alpha\beta}$ as

$$\begin{aligned} P^{\text{triplet}}_{\delta} \;=\; \frac{1}{3!}\sum_{\alpha,\beta,\gamma}\big[&[\mathbb{g}^{\text{triplet}}_{\alpha\beta}\cdot \mathrm{D}_{\delta}\mathbb{x}_{\alpha\beta}+\mathbb{g}^{\text{triplet}}_{\alpha\gamma}\cdot \mathrm{D}_{\delta}\mathbb{x}_{\alpha\gamma}] \\ &+[\mathbb{g}^{\text{triplet}}_{\beta\gamma}\cdot \mathrm{D}_{\delta}\mathbb{x}_{\beta\gamma}+\mathbb{g}^{\text{triplet}}_{\beta\alpha}\cdot \mathrm{D}_{\delta}\mathbb{x}_{\beta\alpha}] \\ &+[\mathbb{g}^{\text{triplet}}_{\gamma\alpha}\cdot \mathrm{D}_{\delta}\mathbb{x}_{\gamma\alpha}+\mathbb{g}^{\text{triplet}}_{\gamma\beta}\cdot \mathrm{D}_{\delta}\mathbb{x}_{\gamma\beta}]\big]. \end{aligned} \tag{67}$$

Likewise, the tetrad-wise contribution to the spatial internal atomistic virtual work expands in terms of the twist-based spatial tetrad interaction force generators $\mathbb{g}^{\text{tetrad}}_{\alpha\beta\gamma}$ as the rather lengthy expression

$$\begin{aligned} P^{\text{tetrad}}_{\delta} \;=\; \frac{1}{4!}\sum_{\alpha,\beta,\gamma,\delta}\big[&[\mathbb{g}^{\text{tetrad}}_{\alpha\delta\beta}\cdot \mathrm{D}_{\delta}\mathbb{x}_{\alpha\delta\beta}+\mathbb{g}^{\text{tetrad}}_{\alpha\delta\gamma}\cdot \mathrm{D}_{\delta}\mathbb{x}_{\alpha\delta\gamma}] \\ &+[\mathbb{g}^{\text{tetrad}}_{\alpha\beta\gamma}\cdot \mathrm{D}_{\delta}\mathbb{x}_{\alpha\beta\gamma}+\mathbb{g}^{\text{tetrad}}_{\alpha\beta\delta}\cdot \mathrm{D}_{\delta}\mathbb{x}_{\alpha\beta\delta}] \\ &+[\mathbb{g}^{\text{tetrad}}_{\alpha\gamma\delta}\cdot \mathrm{D}_{\delta}\mathbb{x}_{\alpha\gamma\delta}+\mathbb{g}^{\text{tetrad}}_{\alpha\gamma\beta}\cdot \mathrm{D}_{\delta}\mathbb{x}_{\alpha\gamma\beta}] \\ &+[\mathbb{g}^{\text{tetrad}}_{\beta\alpha\delta}\cdot \mathrm{D}_{\delta}\mathbb{x}_{\beta\alpha\delta}+\mathbb{g}^{\text{tetrad}}_{\beta\alpha\gamma}\cdot \mathrm{D}_{\delta}\mathbb{x}_{\beta\alpha\gamma}] \\ &+[\mathbb{g}^{\text{tetrad}}_{\beta\delta\gamma}\cdot \mathrm{D}_{\delta}\mathbb{x}_{\beta\delta\gamma}+\mathbb{g}^{\text{tetrad}}_{\beta\delta\alpha}\cdot \mathrm{D}_{\delta}\mathbb{x}_{\beta\delta\alpha}] \\ &+[\mathbb{g}^{\text{tetrad}}_{\beta\gamma\alpha}\cdot \mathrm{D}_{\delta}\mathbb{x}_{\beta\gamma\alpha}+\mathbb{g}^{\text{tetrad}}_{\beta\gamma\delta}\cdot \mathrm{D}_{\delta}\mathbb{x}_{\beta\gamma\delta}] \\ &+[\mathbb{g}^{\text{tetrad}}_{\gamma\beta\delta}\cdot \mathrm{D}_{\delta}\mathbb{x}_{\gamma\beta\delta}+\mathbb{g}^{\text{tetrad}}_{\gamma\beta\alpha}\cdot \mathrm{D}_{\delta}\mathbb{x}_{\gamma\beta\alpha}] \\ &+[\mathbb{g}^{\text{tetrad}}_{\gamma\delta\alpha}\cdot \mathrm{D}_{\delta}\mathbb{x}_{\gamma\delta\alpha}+\mathbb{g}^{\text{tetrad}}_{\gamma\delta\beta}\cdot \mathrm{D}_{\delta}\mathbb{x}_{\gamma\delta\beta}] \\ &+[\mathbb{g}^{\text{tetrad}}_{\gamma\alpha\beta}\cdot \mathrm{D}_{\delta}\mathbb{x}_{\gamma\alpha\beta}+\mathbb{g}^{\text{tetrad}}_{\gamma\alpha\delta}\cdot \mathrm{D}_{\delta}\mathbb{x}_{\gamma\alpha\delta}] \\ &+[\mathbb{g}^{\text{tetrad}}_{\delta\gamma\beta}\cdot \mathrm{D}_{\delta}\mathbb{x}_{\delta\gamma\beta}+\mathbb{g}^{\text{tetrad}}_{\delta\gamma\alpha}\cdot \mathrm{D}_{\delta}\mathbb{x}_{\delta\gamma\alpha}] \\ &+[\mathbb{g}^{\text{tetrad}}_{\delta\beta\alpha}\cdot \mathrm{D}_{\delta}\mathbb{x}_{\delta\beta\alpha}+\mathbb{g}^{\text{tetrad}}_{\delta\beta\gamma}\cdot \mathrm{D}_{\delta}\mathbb{x}_{\delta\beta\gamma}] \\ &+[\mathbb{g}^{\text{tetrad}}_{\delta\alpha\gamma}\cdot \mathrm{D}_{\delta}\mathbb{x}_{\delta\alpha\gamma}+\mathbb{g}^{\text{tetrad}}_{\delta\alpha\beta}\cdot \mathrm{D}_{\delta}\mathbb{x}_{\delta\alpha\beta}]\big]. \end{aligned} \tag{68}$$

Then, extensive manipulations (detailed in Appendix B.4) demonstrate that the principle of spatial atomistic virtual work proves equivalent to the spatial atomistic equilibrium condition $\mathbb{k}^{\text{int}}_{\alpha}+\mathbb{k}^{\text{ext}}_{\alpha}=\mathbb{0}$ with $\mathbb{k}^{\text{int}}_{\alpha}:=\mathbb{k}^{\text{pair}}_{\alpha}+\mathbb{k}^{\text{triplet}}_{\alpha}+\mathbb{k}^{\text{tetrad}}_{\alpha}$, whereby the twist-based spatial triplet and tetrad interaction forces follow in the already established fashion from the corresponding interaction force generators. □

Remark. For a dimensional reduction to a *shell* (bending stiffness only) with the atomistic positions constrained to a two-dimensional manifold, the twist-based spatial tetrad interaction forces degenerate to

$$\mathbb{k}^{\text{tetrad}}_{\alpha\beta\gamma\delta}=2\big[\mathbb{x}_{\beta\gamma}\times[\mathbb{g}^{\text{tetrad}}_{[\alpha\beta]\gamma}+\mathbb{g}^{\text{tetrad}}_{[\gamma\alpha]\beta}]+\mathbb{x}_{\gamma\delta}\times[\mathbb{g}^{\text{tetrad}}_{[\alpha\gamma]\delta}+\mathbb{g}^{\text{tetrad}}_{[\delta\alpha]\gamma}]+\mathbb{x}_{\delta\beta}\times[\mathbb{g}^{\text{tetrad}}_{[\alpha\delta]\beta}+\mathbb{g}^{\text{tetrad}}_{[\beta\alpha]\delta}]\big].\quad\square$$

Remark. For a dimensional reduction to a *beam* (bending stiffness only) with the atomistic positions constrained to a one-dimensional manifold, the twist-based spatial triplet interaction forces degenerate to

$$\mathbb{k}^{\text{triplet}}_{\alpha\beta\gamma}=\mathbb{g}^{\text{triplet}}_{\alpha\beta}+\mathbb{g}^{\text{triplet}}_{\alpha\gamma}\quad\text{with}\quad\mathbb{k}^{\text{tetrad}}_{\alpha\beta\gamma\delta}\equiv\mathbb{0}.\quad\square$$

Remark. For a dimensional reduction to a *shaft* (torsion stiffness only) with the atomistic positions constrained to a one-dimensional manifold, the twist-based spatial tetrad interaction forces degenerate to

$$\mathbb{k}^{\text{tetrad}}_{\alpha\beta\gamma\delta}=2\,\mathbb{x}_{\beta\gamma}\times\mathbb{g}^{\text{tetrad}}_{[\beta\gamma]\alpha}\quad\text{with}\quad\mathbb{k}^{\text{triplet}}_{\alpha\beta\gamma}\equiv\mathbb{0}.\quad\square$$

5.3. Material twist-based parametrisation

For the material setting, we consider the atomistic triplet and tetrad contributions – E^{triplet} and E^{tetrad} – to the internal potential energy E^{int} expanded in terms of their densities – $w_t^{\alpha\beta\gamma}$ and $w_t^{\alpha\beta\gamma\delta}$ – per unit *spatial* angle $\phi_{\alpha\beta\gamma}$ and dihedral angle $\phi_{\alpha\beta\gamma\delta}$, respectively. We parameterise these energy densities in terms of material triplet and tetrad twists $\Omega_{\alpha\beta\gamma}$ and $\Omega_{\alpha\beta\gamma\delta}$, respectively, thus

$$E^{\text{triplet}}:=\frac{1}{3!}\sum_{\alpha,\beta,\gamma}\Big[\;w_t^{\alpha\beta\gamma}\,(\Omega_{\alpha\beta\gamma}\,)\,\phi_{\alpha\beta\gamma}\;+w_t^{\beta\gamma\alpha}\,(\Omega_{\beta\gamma\alpha}\,)\,\phi_{\beta\gamma\alpha}\;+w_t^{\gamma\alpha\beta}\,(\Omega_{\gamma\alpha\beta}\,)\,\phi_{\gamma\alpha\beta}\;\Big] \tag{69}$$

$$\begin{aligned} E^{\text{tetrad}}:=\frac{1}{4!}\sum_{\alpha,\beta,\gamma,\delta}\Big[\;&w_t^{\alpha\delta\beta\gamma}(\Omega_{\alpha\delta\beta\gamma})\,\phi_{\alpha\delta\beta\gamma}+w_t^{\alpha\beta\gamma\delta}(\Omega_{\alpha\beta\gamma\delta})\,\phi_{\alpha\beta\gamma\delta}+w_t^{\alpha\gamma\delta\beta}(\Omega_{\alpha\gamma\delta\beta})\,\phi_{\alpha\gamma\delta\beta} \\ &+w_t^{\beta\alpha\delta\gamma}(\Omega_{\beta\alpha\delta\gamma})\,\phi_{\beta\alpha\delta\gamma}+w_t^{\beta\delta\gamma\alpha}(\Omega_{\beta\delta\gamma\alpha})\,\phi_{\beta\delta\gamma\alpha}+w_t^{\beta\gamma\alpha\delta}(\Omega_{\beta\gamma\alpha\delta})\,\phi_{\beta\gamma\alpha\delta} \\ &+w_t^{\gamma\beta\delta\alpha}(\Omega_{\gamma\beta\delta\alpha})\,\phi_{\gamma\beta\delta\alpha}+w_t^{\gamma\delta\alpha\beta}(\Omega_{\gamma\delta\alpha\beta})\,\phi_{\gamma\delta\alpha\beta}+w_t^{\gamma\alpha\beta\delta}(\Omega_{\gamma\alpha\beta\delta})\,\phi_{\gamma\alpha\beta\delta} \end{aligned}$$

$$+ w_t^{\delta\gamma\beta\alpha}(\Omega_{\delta\gamma\beta\alpha})\,\phi_{\delta\gamma\beta\alpha} + w_t^{\delta\beta\alpha\gamma}(\Omega_{\delta\beta\alpha\gamma})\,\phi_{\delta\beta\alpha\gamma} + w_t^{\delta\alpha\gamma\beta}(\Omega_{\delta\alpha\gamma\beta})\,\phi_{\delta\alpha\gamma\beta}\,\big].$$

Observe that the spatial and material potential energy densities relate, for example, as

$$w_t^{\alpha\beta\gamma} = \Omega_{\alpha\beta\gamma}\, w_0^{\alpha\beta\gamma} \quad \text{and} \quad w_t^{\alpha\beta\gamma\delta} = \Omega_{\alpha\beta\gamma\delta}\, w_0^{\alpha\beta\gamma\delta}, \tag{70}$$

whereby the material triplet and tetrad twists $\Omega_{\alpha\beta\gamma}$ and $\Omega_{\alpha\beta\gamma\delta}$, respectively, serve to transform material into spatial densities.

5.4. Material twist-based interaction forces

The (resultant) material atomistic forces corresponding to the twist-based triplet and tetrad potentials expand, as before, as sums of the corresponding material interaction forces:

$$\mathbb{K}_\alpha^{\text{triplet}} = \frac{1}{2!}\sum_{\beta,\gamma}\mathbb{K}_{\alpha\beta\gamma}^{\text{triplet}} \quad \text{and} \quad \mathbb{K}_\alpha^{\text{tetrad}} = \frac{1}{3!}\sum_{\beta,\gamma,\delta}\mathbb{K}_{\alpha\beta\gamma\delta}^{\text{tetrad}}, \tag{71}$$

with the twist-based material triplet and tetrad interaction forces given by

$$\begin{aligned}
\mathbb{K}_{\alpha\beta\gamma}^{\text{triplet}} &:= -\frac{\partial w_t^{\alpha\beta\gamma}}{\partial \mathbb{X}_\alpha}\,\phi_{\alpha\beta\gamma} - \frac{\partial w_t^{\beta\gamma\alpha}}{\partial \mathbb{X}_\alpha}\,\phi_{\beta\gamma\alpha} - \frac{\partial w_t^{\gamma\alpha\beta}}{\partial \mathbb{X}_\alpha}\,\phi_{\gamma\alpha\beta}\,, \\
\mathbb{K}_{\alpha\beta\gamma\delta}^{\text{tetrad}} &:= -\frac{\partial w_t^{\alpha\delta\beta\gamma}}{\partial \mathbb{X}_\alpha}\,\phi_{\alpha\delta\beta\gamma} - \frac{\partial w_t^{\alpha\beta\gamma\delta}}{\partial \mathbb{X}_\alpha}\,\phi_{\alpha\beta\gamma\delta} - \frac{\partial w_t^{\alpha\gamma\delta\beta}}{\partial \mathbb{X}_\alpha}\,\phi_{\alpha\gamma\delta\beta} \\
&\quad - \frac{\partial w_t^{\beta\alpha\delta\gamma}}{\partial \mathbb{X}_\alpha}\,\phi_{\beta\alpha\delta\gamma} - \frac{\partial w_t^{\beta\delta\gamma\alpha}}{\partial \mathbb{X}_\alpha}\,\phi_{\beta\delta\gamma\alpha} - \frac{\partial w_t^{\beta\gamma\alpha\delta}}{\partial \mathbb{X}_\alpha}\,\phi_{\beta\gamma\alpha\delta} \\
&\quad - \frac{\partial w_t^{\gamma\beta\delta\alpha}}{\partial \mathbb{X}_\alpha}\,\phi_{\gamma\beta\delta\alpha} - \frac{\partial w_t^{\gamma\delta\alpha\beta}}{\partial \mathbb{X}_\alpha}\,\phi_{\gamma\delta\alpha\beta} - \frac{\partial w_t^{\gamma\alpha\beta\delta}}{\partial \mathbb{X}_\alpha}\,\phi_{\gamma\alpha\beta\delta} \\
&\quad - \frac{\partial w_t^{\delta\gamma\beta\alpha}}{\partial \mathbb{X}_\alpha}\,\phi_{\delta\gamma\beta\alpha} - \frac{\partial w_t^{\delta\beta\alpha\gamma}}{\partial \mathbb{X}_\alpha}\,\phi_{\delta\beta\alpha\gamma} - \frac{\partial w_t^{\delta\alpha\gamma\beta}}{\partial \mathbb{X}_\alpha}\,\phi_{\delta\alpha\gamma\beta}.
\end{aligned} \tag{72}$$

Manipulations analogous to those for the spatial interaction forces provided in Appendix B.1 lead to concise expressions in terms of twist-based material triplet and tetrad interaction force generators $\mathbb{G}_{\alpha\beta}^{\text{triplet}}$ and $\mathbb{G}_{\alpha\beta\gamma}^{\text{tetrad}}$ as

$$\begin{aligned}
\mathbb{K}_{\alpha\beta\gamma}^{\text{triplet}} &:= 2\big[\mathbb{G}_{[\alpha\beta]}^{\text{triplet}} + \mathbb{G}_{[\alpha\gamma]}^{\text{triplet}}\big], \\
\mathbb{K}_{\alpha\beta\gamma\delta}^{\text{tetrad}} &:= 2\big[\mathbb{X}_{\beta\gamma} \times [\mathbb{G}_{[\alpha\beta]\gamma}^{\text{tetrad}} + \mathbb{G}_{[\gamma\alpha]\beta}^{\text{tetrad}} + \mathbb{G}_{[\beta\gamma]\alpha}^{\text{tetrad}}] \\
&\quad + \mathbb{X}_{\gamma\delta} \times [\mathbb{G}_{[\alpha\gamma]\delta}^{\text{tetrad}} + \mathbb{G}_{[\delta\alpha]\gamma}^{\text{tetrad}} + \mathbb{G}_{[\gamma\delta]\alpha}^{\text{tetrad}}] \\
&\quad + \mathbb{X}_{\delta\beta} \times [\mathbb{G}_{[\alpha\delta]\beta}^{\text{tetrad}} + \mathbb{G}_{[\beta\alpha]\delta}^{\text{tetrad}} + \mathbb{G}_{[\delta\beta]\alpha}^{\text{tetrad}}]\big].
\end{aligned} \tag{73}$$

Likewise in analogy to the spatial interaction force generators, the twist-based material triplet and tetrad interaction force generators follow as the pair- and triplet-wise sensitivities of the twist-based triplet and tetrad potentials. The complete sets of these would read in analogy to their spatial counterparts provided in Appendix B.2; the following are but two examples

$$\begin{aligned}
\mathbb{G}_{\alpha\beta}^{\text{triplet}} &:= \frac{\partial w_t^{\alpha\beta\gamma}}{\partial \mathbb{X}_{\alpha\beta}}\,\phi_{\alpha\beta\gamma} \\
&=: K_{\alpha\beta\gamma}^{\text{triplet}}\;\; \mathbb{P}_{\alpha\beta}^{\perp}\cdot\mathbb{N}_{\alpha\gamma}
\end{aligned} \quad \text{and} \quad
\begin{aligned}
\mathbb{G}_{\alpha\beta\gamma}^{\text{tetrad}} &:= \frac{\partial w_t^{\alpha\beta\gamma\delta}}{\partial \mathbb{X}_{\alpha\beta\gamma}}\,\phi_{\alpha\beta\gamma\delta} \\
&=: K_{\alpha\beta\gamma\delta}^{\text{tetrad}}\;\; \mathbb{P}_{\alpha\beta\gamma}^{\perp}\cdot\mathbb{N}_{\alpha\beta\delta}
\end{aligned}. \tag{74}$$

Therein, we also introduced the signed "magnitudes" of the twist-based material triplet and tetrad interaction force generators. The complete set would read in analogy to its spatial counterpart provided in Appendix B.2; the following are but two examples

$$K_{\alpha\beta\gamma}^{\text{triplet}} := \frac{\partial w_t^{\alpha\beta\gamma}}{\partial \Omega_{\alpha\beta\gamma}} \quad \text{and} \quad K_{\alpha\beta\gamma\delta}^{\text{tetrad}} := \frac{\partial w_t^{\alpha\beta\gamma\delta}}{\partial \Omega_{\alpha\beta\gamma\delta}}. \tag{75}$$

It is interesting to observe that these expand in terms of their spatial counterparts for example as

$$K_{\alpha\beta\gamma}^{\text{triplet}} = w_0^{\alpha\beta\gamma} - \omega_{\alpha\beta\gamma}\, k_{\alpha\beta\gamma}^{\text{triplet}} \quad \text{and} \quad K_{\alpha\beta\gamma\delta}^{\text{tetrad}} = w_0^{\alpha\beta\gamma\delta} - \omega_{\alpha\beta\gamma\delta}\, k_{\alpha\beta\gamma\delta}^{\text{tetrad}}. \tag{76}$$

Thus, the features of the classical energy–momentum format of the Eshelby stress established in the continuum setting are apparent in the expressions of the signed "magnitudes" of the twist-based material triplet and tetrad interaction force generators. The noteworthy difference, however, is that in the twist-based atomistic setting energy densities are expressed per unit angle rather than per unit length, area or volume (or per unit volume as in the continuum setting), respectively.

6. Conclusion

Within molecular dynamics or molecular statics two-, three- and four body potentials describe empirically the complex energetic landscape of atomistic systems by considering the energetic interactions between potentially all possible combinations of atomistic

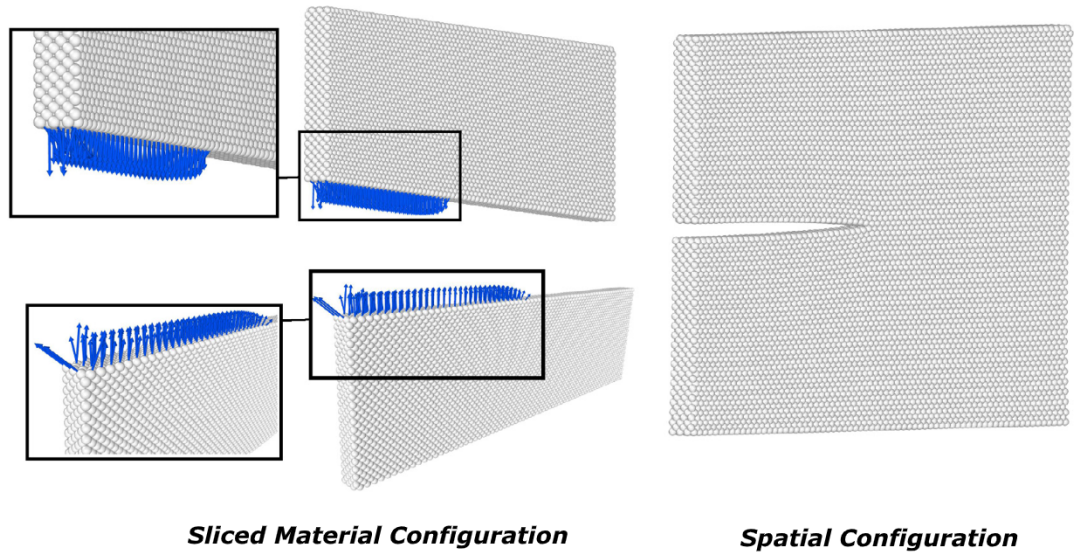


Fig. 17. Crack in an atomistic FCC lattice under vertical Mode I loading. Aluminium is modelled by the Lennard-Jones potential with parameters $\sigma = 2.925$, $\epsilon = 0.17432$ and lattice constant 4.51 Å. Initially, a short pre-crack extending horizontally over only a few lattice spacings is introduced by switching off the interaction between neighbouring atoms. Based on the material-force-based bond deletion criterion from Birang O. and Steinmann (2021), we subsequently evolve the crack by prescribing an increasing displacement to the top and bottom surfaces of the specimen. Atomistic (resultant) material forces showcased by (blue) arrows drive energetically favourable re-organisations of the material atomistic configuration. They also reflect the non-symmetric arrangement of atoms in the FCC lattice relative to the geometry of the finite computational domain.

pairs, triplets and tetrads. Pair potentials are commonly parameterised in terms of atomistic pair lengths, whereas triplet and tetrad potentials are traditionally parameterised in terms of atomistic triplet angles and tetrad dihedral angles, respectively.

This well-established classical approach has been extended here in three ways. First, inspired by continuum kinematics, we proposed triplet areas and tetrad volumes as kinematic measures of three- and four body interactions as an alternative to the well-accepted triplet angles and tetrad dihedral angles. Second, we expressed the atomistic pair, triplet and tetrad contributions to the internal potential energy either in terms of stretches, i.e. the ratios of spatial and material pair lengths, triplet areas and tetrad volumes, or in terms of twists, i.e. the ratios of spatial and material triplet angles and tetrad dihedral angles. Third, we expanded the internal potential energy contributions in terms of pair, triplet and tetrad densities, either per unit pair length, triplet area and tetrad volume, respectively, or per unit triplet angle and tetrad dihedral angle, respectively.

These novel parameterisations prove attractive for four reasons. First, the corresponding spatial atomistic forces can be expressed in a compact format in terms of interaction forces. Their generators and signed magnitudes reduce naturally to dimensionally constrained situations as required for 2d and 1d materials. Second, the corresponding material atomistic forces that drive energetically favourable re-organisations of the material atomistic configuration follow in a straightforward manner, again in terms of interaction forces, their generators and signed magnitudes. Third, in terms of interaction force generators and their signed magnitudes, the material interaction forces display a format structurally identical to their spatial counterparts. Fourth, the signed magnitudes of the spatial and material interaction forces display an energy–momentum, Eshelby-type format as celebrated in the continuum setting of configurational mechanics.

Taken together, we have provided the appropriate framework to consider material or rather configurational atomistic forces in (discrete) atomistic systems characterised by different parameterisations of two-, three- and four body potentials with a focus on studying the tendency of generic atomistic defects to propagate. Fig. 17 highlights a typical computational example of atomistic fracture mechanics based on the prototypical Lennard-Jones two-body potential (see Footnote 5).

Our future work will focus on computational studies of atomistic defect propagation based on concrete selections of atomistic potentials, as for example for silicon or graphene, thereby including the consideration of cut-offs. Moreover, we will elaborate a corresponding twist-based approach to peridynamics.

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Appendix A. Supplementary data

Supplementary material related to this article can be found online at https://doi.org/10.1016/j.jmps.2021.104507.

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