Electronic Supplementary Information (ESI)

Unified approach to multipolar polarisation and charge transfer for ions: microhydrated Na^+

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List of Abbreviations

GF – Global Frame
ILF – Ion Local Frame
WLF – Water Local Frame
NN – Nearest Neighbour
WLF_{NN} – Water Local Frame on Nearest-Neighbour Water Molecule

List of Variables

n – Number of water molecules in the system
d – Number of intermolecular degrees of freedom
A – Number of unbonded atoms in a general system (including ions)
L – Number of rigid linear molecules in a general system
M – Total number of rigid molecules in a general system (including rigid linear molecules)
Ao – Denotes the atom at the origin of a particular coordinate frame
Ax – Denotes the atom whose position defines the x-axis of a particular coordinate frame
Axy - Denotes the atom whose position defines the xy-plane of a particular coordinate frame
R – A general rotation matrix that transforms a vector from one coordinate frame to another
r – An interatomic vector
α_i^A - The i^{th} Cartesian coordinate of atom A in an initial frame, i.e. prior to rotation of the coordinate system
ζ_i^A - The i^{th} Cartesian coordinate of atom A in a target frame, i.e. after rotation of the coordinate system
α, β, γ - Euler angles describing the rotation from an initial to a target coordinate system
χ_1, χ_2 - Rotation angles describing the rotation specifically from the ILF to the WLF_{NN}

Internal Coordinate Definitions

As an example of the general applicability of the method, we describe here a local frame for a heterogeneous system where the intermolecular polarisation is to be included. For a system of *A* atoms and *M* rigid molecules (where *L* of those *M* are linear), the total number of intermolecular degrees of freedom, *d*, in the rigid body approximation is given by [1].

$$d = 6(M-1) - L + 3A \qquad \text{(A1)}$$

For the $Na^+(H_2O)_n$ systems studied in this communication, there are *M*=*n* rigid water molecules (*L*=0 as no linear molecules are present) and *A*=1 atomic ions. The value of *d* for such a system is given by

$$d = 6(n-1) + 3 \qquad \text{(A2)}$$

The coordinates must specify the position of the ion in the system, as well as the position and orientation of each of the *n* rigid water molecules. As the smallest $Na^+(H_2O)_n$ system is composed of one ion and one water molecule, we choose to define the coordinate system using the ion and its Nearest-Neighbour water molecule (denoted NN) for all the systems studied in this work. This coordinate system will be referred to as the Ion Local Frame (ILF). The ion (Na^+) is placed at the origin, with the *x*-axis defined to lie along the vector from the ion to the oxygen atom (O^{NN}) of the nearest water molecule in the system. The lowest numbered hydrogen atom (H_a^{NN}) of the nearest-neighbour water molecule is placed in the *xy*-plane, and the *z*-axis is chosen to form a right-handed coordinate system. Note that the initial numbering is arbitrary and was assigned by the program used to extract geometries from simulation data in this work.

Figure A1 shows an example of the rotation of a $Na^+(H_2O)_2$ system from the GF (initial frame) to the ILF (target frame).

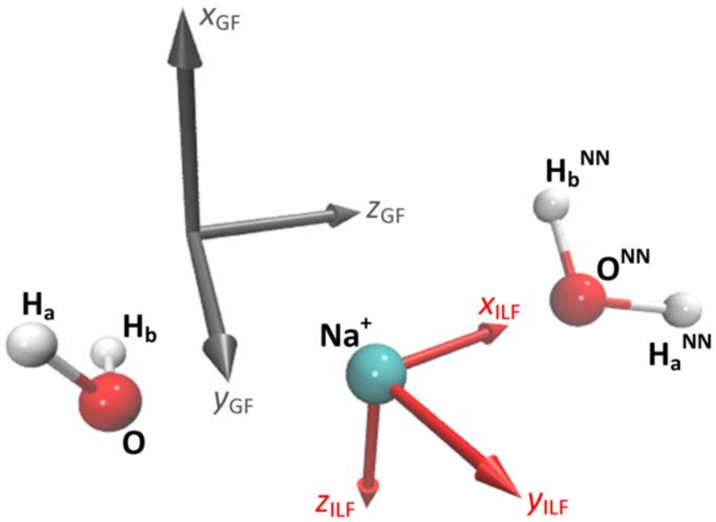

Figure A1 – Relative orientations of the arbitrary global frame (shown in grey) and the ion local frame (shown in red). The origin of the ion local frame is the Na^+ atom, the *x*-axis of the ILF points at the oxygen atom of the nearest neighbour water molecule, O^{NN}, whilst the lowest numbered hydrogen of that same molecule, H_a^{NN}, lies in the *xy*-plane of the ILF. The non-NN water (left) is not involved in the definition of the ion local frame.

The first step in the computation of the internal coordinates is to translate and rotate the system from its arbitrarily oriented and centered initial global frame (GF) to the ILF. The process of rotating between an initial and target coordinate system is made general by first denoting a Cartesian coordinate of a system atom *A* in the initial frame as α_i^A (*i*=x,y,z). The origin, *x*-axis and *xy*-plane defining atoms of the target frame will be labeled *Ao*, *Ax* and *Axy,* respectively. For the specific case of the transformation from the GF to the ILF, for example, we have *Ao*= Na^+, *Ax*= O^{NN} and *Axy*= H_a^{NN}. α_i^A is the i^{th} Cartesian coordinate of atom *A* expressed in the GF. The first row of **R**, the orthogonal matrix that performs a rotation between the original and target coordinate frames, is given by[2]

$$R_{1i} = \frac{\alpha_i^{Ax} - \alpha_i^{Ao}}{\left|\mathbf{r}^{AoAx}\right|} \quad (i=1, 2, 3) \quad \text{(A3)}$$

where $\mathbf{r}^{AoAx}$ is the vector from *Ao* to *Ax* . The second row is given by first defining the vector **c**, where

$$c_i = -\frac{\mathbf{r}^{AoAx} \cdot \mathbf{r}^{AoAxy}}{\mathbf{r}^{AoAx} \cdot \mathbf{r}^{AoAx}}(\alpha_i^{Ax} - \alpha_i^{Ao}) + (\alpha_i^{Axy} - \alpha_i^{Ao}) \quad (i=1, 2, 3) \quad \text{(A4)}$$

and the elements of the second row are given by

$$R_{2i} = \frac{c_i}{\sqrt{\boldsymbol{c}\cdot\boldsymbol{c}}} \qquad (i=1, 2, 3) \qquad \text{(A5)}$$

where $\mathbf{r}^{\mathrm{AoAxy}}$ is the vector from *Ao* to A*xy*. The third row is obtained as the vector product of the first and second rows,

$$\mathbf{R}_3 = \mathbf{R}_1 \times \mathbf{R}_2 \qquad \text{(A6)}$$

It is then possible to rotate the position vector, $\boldsymbol{\alpha}^A$, of an atom *A* from the initial frame to the target coordinate system according to

$$\begin{bmatrix} \zeta_1^A \\ \zeta_2^A \\ \zeta_3^A \end{bmatrix} = \mathbf{R} \begin{bmatrix} \alpha_1^A \\ \alpha_2^A \\ \alpha_3^A \end{bmatrix} \qquad \text{(A7)}$$

where ζ_i^A is a Cartesian coordinate of atom *A* expressed in the target frame.

Three internal coordinates are required to describe the ion and nearest water: one positional coordinate, which gives the point along the ILF *x*-axis where the water oxygen atom lies, and two orientational coordinates that describe the positions of the two hydrogen atoms H_a and H_b. These two orientational coordinates will be described in detail below. We first deal with the more general case of water molecules that are not used to define the ILF.

For systems containing more than one water molecule (i.e. $n \geq 2$), each water molecule that is not used to define the frame (i.e. a water molecule that is *not* the NN of the ion) has its position described by three spherical polar coordinates that give the position of its oxygen atom. Such a water molecule's orientation (i.e. the positions of the hydrogen atoms) is described by three Euler angles. As there are a large number of different definitions for these angles, we describe those employed herein for transparency, following the conventions employed by Su and Coppens[3, 4] For the non-nearest neighbour water molecules we first define a Water Local Frame (WLF), with O at the origin, the lowest numbered hydrogen atom, H_a, defining the *x*-axis and the remaining hydrogen atom, H_b, defining the *xy*-plane. The orthogonal matrix **R** that transforms from the ILF (initial frame) to the WLF (target frame) of a particular non-NN water molecule can be written in terms of the atomic Cartesian coordinates in the ILF as above in Eqs. A3, A5 and A6, and is computed in this manner. The identities of *Ao*, *Ax* and *Axy* change (to O, H_a and H_b, respectively) and α_i^A now denotes an ILF Cartesian coordinate, with ζ_i^A defining a WLF Cartesian coordinate. Figure 2 shows the ILF and the WLF for the $Na^+(H_2O)_2$ example system.

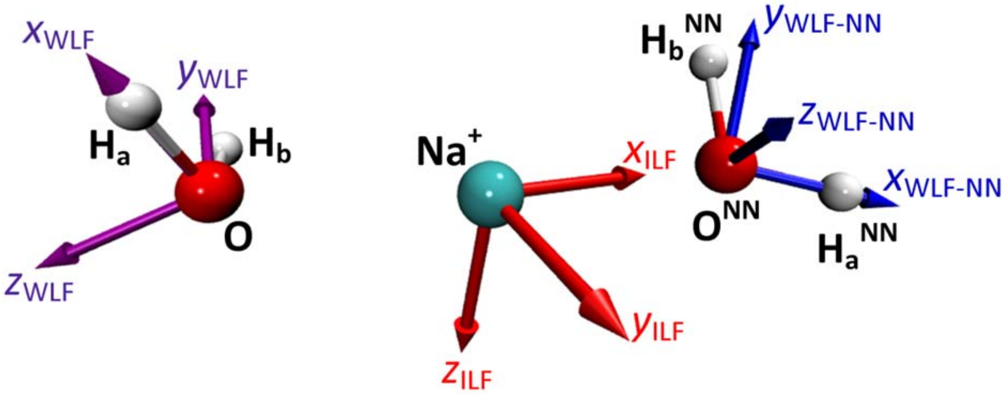

Figure A2 - ILF (red), WLF^{NN} (blue) and WLF (purple) of the $Na^+(H_2O)_2$ system. Each WLF is centred on the oxygen atom of a particular water molecule, the *x*-axis of each WLF points along the O-H_a bond, while the O-H_b bond resides in the *xy*-plane. The *z*-axis is chosen to form a right-handed coordinate system in all three axis systems.

A rotation matrix **R** may alternatively be written in terms of Euler angles α, β and γ, where the transformation is then represented as three successive rotations of firstly α about the initial frame *z*-axis, then β about the initial frame *y*-axis and finally γ about the initial frame *z*-axis, where in each case a positive rotation is counter-clockwise. Each of these individual rotations is associated with one of the following transformation matrices:

$$\mathbf{R}_\alpha = \begin{pmatrix} \cos\alpha & \sin\alpha & 0 \\ -\sin\alpha & \cos\alpha & 0 \\ 0 & 0 & 1 \end{pmatrix} \tag{A8}$$

$$\mathbf{R}_\beta = \begin{pmatrix} \cos\beta & 0 & -\sin\beta \\ 0 & 1 & 0 \\ \sin\beta & 0 & \cos\beta \end{pmatrix} \tag{A9}$$

$$\mathbf{R}_\gamma = \begin{pmatrix} \cos\gamma & \sin\gamma & 0 \\ -\sin\gamma & \cos\gamma & 0 \\ 0 & 0 & 1 \end{pmatrix} \tag{A10}$$

The complete transformation matrix **R** is given by the matrix product $\mathbf{R}_\gamma\mathbf{R}_\beta\mathbf{R}_\alpha$[3, 4]

$$\mathbf{R} = \mathbf{R}_\gamma\mathbf{R}_\beta\mathbf{R}_\alpha = \begin{pmatrix} \cos\alpha\cos\beta\cos\gamma - \sin\alpha\sin\gamma & \sin\alpha\cos\beta\cos\gamma + \cos\alpha\sin\gamma & -\sin\beta\cos\gamma \\ -\cos\alpha\cos\beta\sin\gamma - \sin\alpha\cos\gamma & -\sin\alpha\cos\beta\sin\gamma + \cos\alpha\cos\gamma & \sin\beta\sin\gamma \\ \cos\alpha\sin\beta & \sin\alpha\sin\beta & \cos\beta \end{pmatrix} \tag{A11}$$

We can compute the elements of this matrix from the atomic Cartesian coordinates as above (Eqs. A3, A4 and A5) and subsequently determine the Euler angles by writing them in terms of the elements of **R** as follows. We note here that the reverse transformation (i.e. from the target frame to the initial frame) also depends only on these three coordinates and involves the transpose of **R**, where $\mathbf{R}^\mathrm{T} = \mathbf{R}_\alpha^\mathrm{T}\mathbf{R}_\beta^\mathrm{T}\mathbf{R}_\gamma^\mathrm{T} = \mathbf{R}_{-\alpha}\mathbf{R}_{-\beta}\mathbf{R}_{-\gamma}$. That is, the transformation from the target frame back to the initial frame involves first a rotation about the initial frame *z*-axis by an angle $-\gamma$ (corresponding to a clockwise rotation by an angle γ), followed by a rotation about the initial frame y-axis by an angle $-\beta$ (corresponding to a clockwise rotation by an angle β), then finally a rotation about the initial frame *z*-axis by an angle $-\alpha$ (corresponding to a clockwise rotation by an angle α).

We now calculate the three Euler angles, starting with β, for the case where $R_{33} \neq \pm 1$. From R_{33} in Eq. A11 we have

$$\beta = \arccos(R_{33}) \qquad \text{(A12)}$$

The angle β has a range of [0,π] radians. A result in this range is guaranteed by the arccosine function, which returns a value in the same range. The angles *α* and *γ* have a range of [0,2π] and so require some additional treatment in order to determine their exact values.

Given the value of β computed with eq. A12, we can use either R_{31} or R_{32} to compute α without needing to know *γ*. We will employ R_{31}, where the arccosine function will return a value in the range [0,π] (shown in green in Fig. A3), which covers only half of the range of values the angle can take (marked by a dashed black outline on Fig. A3).

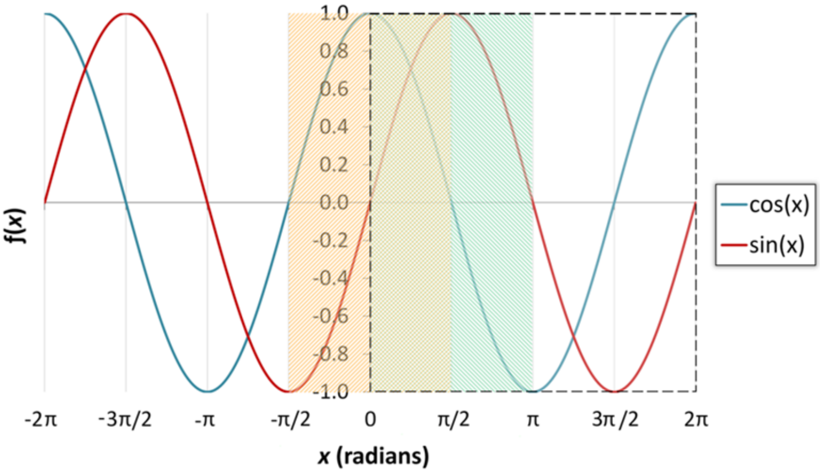

Figure A3 – Regions of the cosine (blue) and sine (red) curves used to determine the correct values of the Euler angles α and γ. Their range [0,2π] is marked with a dashed black outline. The range of values returned by the arccosine function, or [0, π], is marked in green, and the arcsine range, or [-π/2, π/2], is marked in orange.

In order to determine the correct value within the range [0,2π], we need to inspect the sine of *α*, where R_{32} can be used to obtain this value. If positive, *α* is in the range [0,π] and if negative, α is in the range [-π,0] = [π,2π] and the arccosine result must be corrected by subtracting the raw arccosine value from 2π. This can be written more succinctly as

$$\alpha = \begin{cases} \arccos(R_{31}/\sin\beta) & \text{if } R_{32}/\sin\beta \geq 0 \\ 2\pi - \arccos(R_{31}/\sin\beta) & \text{if } R_{32}/\sin\beta < 0 \end{cases} \qquad \text{(A13)}$$

The angle *γ* lies in the same range as *α*, [0,2π]. Again using the value of β, we can compute the angle independently of α using R_{13}, and subsequently determine the corrected value by evaluating the sine of *γ* from R_{23}. The resulting expression is

$$\gamma = \begin{cases} \arccos(-R_{13}/\sin\beta) & \text{if } R_{23}/\sin\beta \geq 0 \\ 2\pi - \arccos(-R_{13}/\sin\beta) & \text{if } R_{23}/\sin\beta < 0 \end{cases} \qquad \text{(A14)}$$

We stated above that the equations given for α and β are only valid if $R_{33} \neq \pm 1$. If this condition is not satisfied, that is $R_{33} = \pm 1$, then the rotation can be described by a single angle, either α or γ. We use α, setting γ=0. The form of the individual rotation matrix for the angle α depends on the specific value of β. However, the value of α can be computed in either case using eq. A16. Again the range of α is [0,2π] and the actual value is determined using sin α. We have that $\cos\beta = R_{33} = \pm 1$, and following from this, that $\sin\beta = 0$. Having set $\gamma = 0$, we also have $\cos\gamma = 1$ and $\sin\gamma = 0$. Substitution into eq. A11 gives

$$\mathbf{R} = \begin{pmatrix} \pm\cos\alpha & \pm\sin\alpha & 0 \\ -\sin\alpha & \cos\alpha & 0 \\ 0 & 0 & \pm 1 \end{pmatrix} \qquad \text{(A15)}$$

which is a matrix describing first a rotation of α radians about the initial frame *z*-axis, followed by a rotation of either 0 or π radians about the initial frame *y*-axis. That is, Eq. A15 can be decomposed into two matrices as follows

$$\mathbf{R} = \mathbf{R}_\beta \mathbf{R}_\alpha = \begin{pmatrix} \pm 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & \pm 1 \end{pmatrix} \begin{pmatrix} \cos\alpha & \sin\alpha & 0 \\ -\sin\alpha & \cos\alpha & 0 \\ 0 & 0 & 1 \end{pmatrix} \qquad \text{(A16)}$$

We can write an expression for α when $R_{33} = \pm 1$ by first writing $\cos\alpha = R_{11}/R_{33}$ and $\sin\alpha = R_{12}/R_{33}$ by consideration of Eq. A15. By the same considerations used above to put the result into the correct range ($0 \leq \alpha \leq 2\pi$) we can then write

$$\alpha = \begin{cases} \arccos(R_{11}/R_{33}) & \text{if } R_{12}/R_{33} \geq 0 \\ 2\pi - \arccos(R_{11}/R_{33}) & \text{if } R_{12}/R_{33} < 0 \end{cases} \qquad \text{(A17)}$$

Using the angles determined with Eqs. A12, A13 and A14 (or in the described special case associated with $R_{33} = \pm 1$) we can construct the individual rotation matrices. The stepwise rotations of the ILF required to transform the coordinate system to the WLF are shown in Fig. A4.

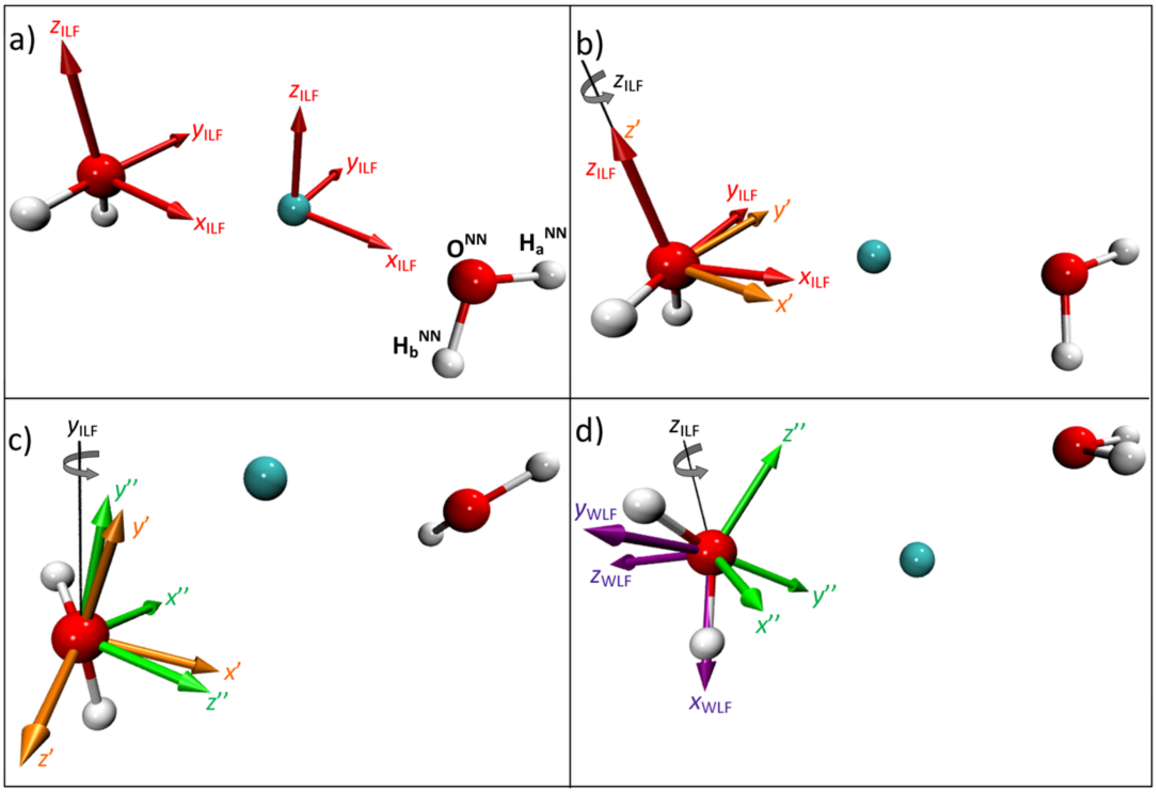

Figure A4 – Rotations of *α*, *β* and *γ* about the *z*, *y* and *z* axes that transform the ILF to the WLF of a non-nearest neighbour water molecule. (a) The ILF (red) is translated from the Na+ atom to O for the sake of clarity. (b) The ILF is rotated through γ=-6.0 radians (or 16^0 clockwise) about the ILF *z*-axis to give the first step in the rotation (orange). (c) The rotated coordinate system (orange) is rotated through β=-1.1 radians (or 297^0 clockwise) about the ILF *x*-axis to give the second step (green). (d) The rotation is completed by rotating through α=-3.3 radians (171^0 clockwise) about the ILF *z*-axis. The direction of rotation is shown with a grey arrow and the axis of rotation for each step is shown in black.

The angles *α*, *β* and *γ* constitute the orientational coordinates for a non-NN water molecule. For the NN water molecule a different approach is required because only two angles are needed to specify its orientation, and so adopting the same approach as for the non-nearest neighbour water molecules results in one redundant coordinate in the description of the system. As a consequence of the definition of the ILF, H_a^{NN} is confined to the ILF *xy*-plane. This is because *Axy* of the ILF (being the lowest numbered hydrogen atom attached to the NN water molecule of the ion) is the same as *Ax* of the WLF of the NN water molecule (being the lowest numbered hydrogen attached to that water molecule), so the initial and target coordinate systems are already partially aligned. This relationship is illustrated in Fig. A2, where the ILF and WLF_{NN} are shown. The orientation of the NN water molecule is restricted. It is free to rotate about both the WLF^{NN} *z*-axis and the confined O^{NN}-H_a^{NN} bond. The angles of these rotations constitute the two orientational coordinates for the NN water molecule. The rotation can be decomposed into two successive rotations (cf. three for a non-NN): first by an angle χ_1 about the ILF *z*-axis and second by an angle χ_2 about the ILF *x*-axis. A positive rotation is again counter-clockwise, and the two rotations can be described by the following matrices

$$\mathbf{R}_{\chi_1} = \begin{pmatrix} \cos\chi_1 & \sin\chi_1 & 0 \\ -\sin\chi_1 & \cos\chi_1 & 0 \\ 0 & 0 & 1 \end{pmatrix} \qquad \text{(A18)}$$

$$\mathbf{R}_{\chi_2} = \begin{pmatrix} 1 & 0 & 0 \\ 0 & \cos\chi_2 & \sin\chi_2 \\ 0 & -\sin\chi_2 & \cos\chi_2 \end{pmatrix} \qquad \text{(A19)}$$

The full rotation matrix from the initial (ILF) to the target (WLF^{NN}) coordinate system is again given by the product of the individual rotation matrices

$$\mathbf{R} = \mathbf{R}_{\chi_2}\mathbf{R}_{\chi_1} = \begin{pmatrix} \cos\chi_1 & \sin\chi_1 & 0 \\ -\sin\chi_1\cos\chi_2 & \cos\chi_1\cos\chi_2 & \sin\chi_2 \\ \sin\chi_1\sin\chi_2 & -\cos\chi_1\sin\chi_2 & \cos\chi_2 \end{pmatrix} \qquad \text{(A20)}$$

The range of the angles χ_1 and χ_2 is [0,2π] and again we can use the sine of each angle to determine their values as shown in Figure A3. The angles are given in terms of the elements of the full rotation matrix in Eq. A20 by

$$\chi_1 = \begin{cases} \arccos(R_{11}) & \text{if } R_{12} \geq 0 \\ 2\pi - \arccos(R_{11}) & \text{if } R_{12} < 0 \end{cases} \qquad \text{(A21)}$$

$$\chi_2 = \begin{cases} \arccos(R_{33}) & \text{if } R_{23} \geq 0 \\ 2\pi - \arccos(R_{33}) & \text{if } R_{23} < 0 \end{cases} \qquad \text{(A22)}$$

χ_1 is the angle between the confined O^{NN}-H$_a^{NN}$ bond vector and the ILF *x*-axis and χ_2 is the angle between the *z*-axes of the ILF and WLF^{NN}.

Figure A5 shows the successive rotations of χ_1 and χ_2 about the ILF *z* and *x*-axes, respectively, for the example $Na^+(H_2O)_2$ system that transform between the ILF and WLF^{NN}. The angles χ_1 and χ_2 constitute the orientational coordinates of the NN water.

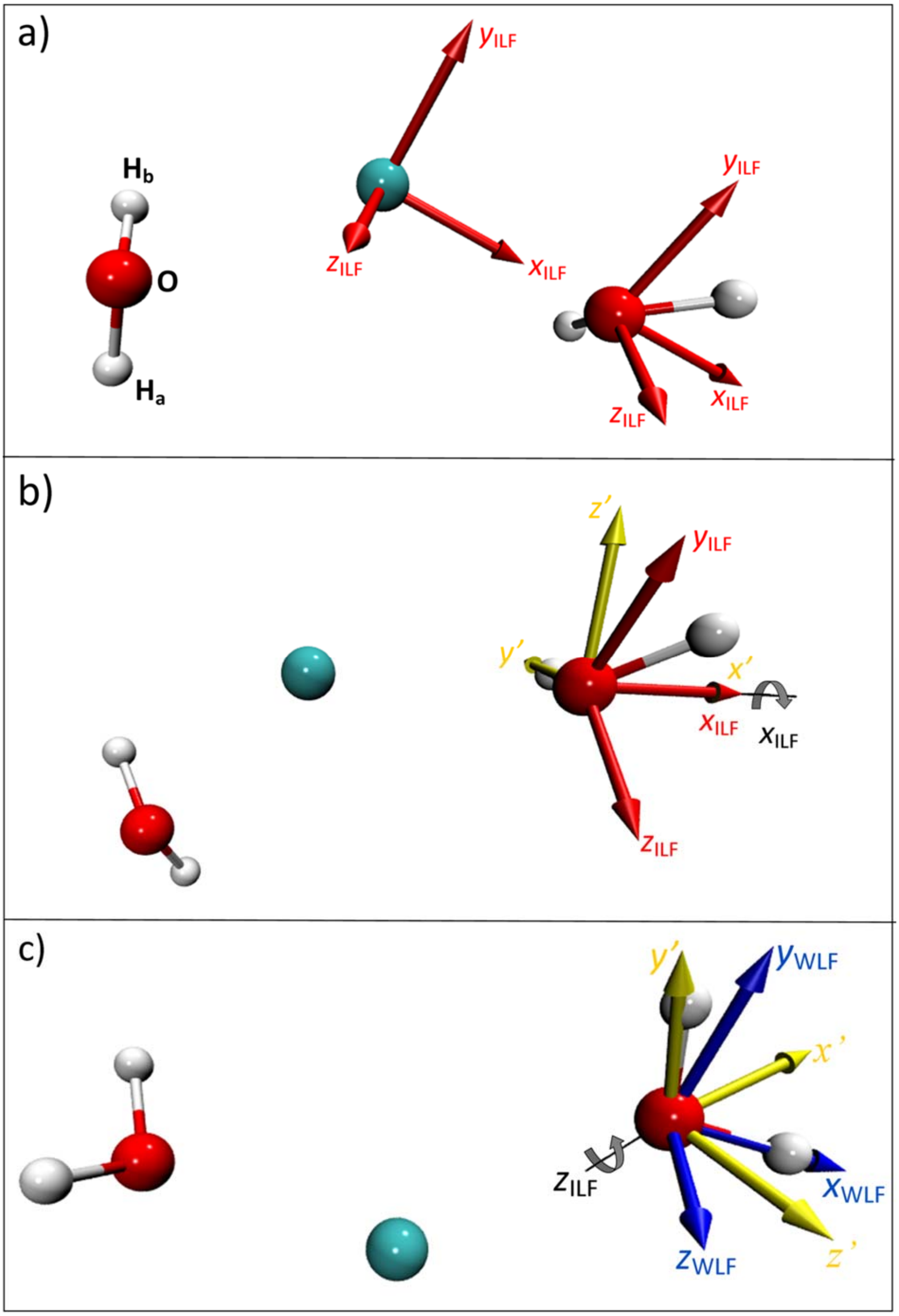

Figure A5 – Rotations that move the system from the ILF to the WLF. (a) The ILF (red) is purely translated to the nearest-neighbour water molecule. (b) The system is rotated through χ_2 = -4.1 radians (165^0 clockwise) about the ILF *x*-axis to give the first step in the rotation (yellow). (c) The system is rotated through χ_1 = -0.7 radians (320^0 clockwise) about the ILF *z*-axis to give the WLF^{NN}. The direction of rotation is shown with a grey arrow and the axis of rotation is shown in black in each case.

In summary, *r*, χ_1 and χ_2 are sufficient to describe the position and orientation of the NN water molecule, while each of the *n*-1 non-NN water molecules requires the six coordinates $r, \theta, \varphi, \alpha, \beta, \gamma$. In both cases, *r* is the distance between the ion and the oxygen atom of a water molecule.

$$r^2 = \sum_{i=1}^{3} (\alpha_i^0 - \alpha_i^{ion})^2 \qquad \text{(A23)}$$

The resulting 3+6(*n*-1) system coordinates can be used to build Kriging models for the AMMs of the atomic ion.

References

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Tables of AMM Prediction Results

Table 1. Average absolute atomic multipole moment errors across the test set geometries for the 25 multipole moments of Na^+ in all six $Na^+(H_2O)_n$ water clusters. The average absolute error is calculated from

$$\mathrm{AAE} = \frac{1}{N}\sum_{i=1}^{N}|Q_{\ell m} - \bar{Q}_{\ell m}|$$

where $\bar{Q}_{\ell m}$ is a predicted AMM, $Q_{\ell m}$ is the true value of the AMM and *N* is the number of test examples.

n	1	2	3	4	5	6
AMM						
Q_{00}	1.78E-05	1.66E-03	2.78E-03	3.06E-03	2.72E-03	2.58E-03
Q_{10}	1.95E-03	2.35E-03	3.24E-03	3.43E-03	3.77E-03	3.57E-03
Q_{11c}	1.96E-03	2.30E-03	3.23E-03	3.59E-03	3.94E-03	3.82E-03
Q_{11s}	1.86E-03	2.24E-03	3.05E-03	3.56E-03	3.67E-03	3.73E-03
Q_{20}	1.36E-03	1.88E-03	2.97E-03	3.77E-03	4.15E-03	4.01E-03
Q_{21c}	1.42E-03	1.88E-03	2.89E-03	3.82E-03	4.00E-03	4.13E-03
Q_{21s}	1.37E-03	1.88E-03	2.86E-03	3.92E-03	4.24E-03	4.30E-03
Q_{22c}	1.40E-03	1.89E-03	2.82E-03	3.66E-03	4.12E-03	4.26E-03
Q_{22s}	1.40E-03	1.90E-03	2.88E-03	3.90E-03	4.30E-03	4.01E-03
Q_{30}	3.60E-03	4.98E-03	8.11E-03	1.31E-02	1.84E-02	1.90E-02
Q_{31c}	3.53E-03	5.20E-03	7.97E-03	1.31E-02	1.85E-02	1.92E-02
Q_{31s}	3.82E-03	5.03E-03	8.15E-03	1.34E-02	1.82E-02	1.96E-02
Q_{32c}	4.11E-03	5.31E-03	8.39E-03	1.36E-02	1.75E-02	1.92E-02
Q_{32s}	3.85E-03	5.20E-03	8.07E-03	1.30E-02	1.88E-02	1.90E-02
Q_{33c}	3.59E-03	4.77E-03	7.99E-03	1.31E-02	1.96E-02	1.86E-02
Q_{33s}	3.67E-03	4.84E-03	7.72E-03	1.35E-02	1.94E-02	1.92E-02
Q_{40}	2.54E-03	8.96E-03	1.74E-02	2.84E-02	4.71E-02	5.47E-02
Q_{41c}	2.60E-03	9.52E-03	1.69E-02	2.83E-02	4.68E-02	5.75E-02
Q_{41s}	2.62E-03	8.95E-03	1.75E-02	2.86E-02	4.63E-02	5.57E-02
Q_{42c}	2.75E-03	8.92E-03	1.75E-02	3.01E-02	4.65E-02	5.45E-02
Q_{42s}	2.49E-03	9.30E-03	1.72E-02	2.98E-02	4.77E-02	5.47E-02
Q_{43c}	2.69E-03	9.39E-03	1.76E-02	2.85E-02	4.52E-02	5.64E-02
Q_{43s}	2.76E-03	9.78E-03	1.73E-02	3.22E-02	4.59E-02	5.62E-02
Q_{44c}	2.60E-03	9.01E-03	1.69E-02	2.98E-02	4.69E-02	5.46E-02
Q_{44s}	2.64E-03	8.88E-03	1.72E-02	2.97E-02	4.86E-02	5.63E-02

Table 2. Maximum absolute atomic multipole moment errors across the test set geometries for the 25 multipole moments of Na$^+$ modelled in all six Na$^+$…water clusters. The maximum absolute error is the largest value of $|Q_{\ell m} - \bar{Q}_{\ell m}|$ found in the test set.

n	1	2	3	4	5	6
AMM						
Q_{00}	3.37E-04	8.89E-03	1.55E-02	1.53E-02	1.27E-02	1.41E-02
Q_{10}	1.29E-02	1.52E-02	1.72E-02	1.62E-02	2.00E-02	1.85E-02
Q_{11c}	7.72E-03	1.29E-02	1.72E-02	1.91E-02	1.68E-02	1.83E-02
Q_{11s}	7.93E-03	1.20E-02	1.69E-02	1.66E-02	2.62E-02	2.45E-02
Q_{20}	7.02E-03	1.16E-02	1.17E-02	1.76E-02	2.28E-02	2.12E-02
Q_{21c}	7.28E-03	1.21E-02	2.11E-02	2.01E-02	2.14E-02	1.93E-02
Q_{21s}	8.59E-03	1.15E-02	1.27E-02	1.74E-02	1.90E-02	2.18E-02
Q_{22c}	7.03E-03	8.59E-03	1.34E-02	2.16E-02	2.11E-02	1.77E-02
Q_{22s}	8.21E-03	9.48E-03	1.32E-02	1.97E-02	2.03E-02	1.67E-02
Q_{30}	2.09E-02	2.88E-02	4.96E-02	5.90E-02	8.67E-02	8.33E-02
Q_{31c}	2.07E-02	3.54E-02	7.08E-02	5.90E-02	7.47E-02	8.52E-02
Q_{31s}	2.20E-02	2.96E-02	3.77E-02	8.10E-02	7.69E-02	7.52E-02
Q_{32c}	2.22E-02	4.74E-02	1.01E-01	8.19E-02	1.03E-01	7.89E-02
Q_{32s}	2.37E-02	2.91E-02	5.34E-02	7.42E-02	8.47E-02	8.70E-02
Q_{33c}	1.89E-02	4.46E-02	5.47E-02	7.24E-02	1.26E-01	9.54E-02
Q_{33s}	2.29E-02	2.41E-02	4.35E-02	1.56E-01	1.07E-01	1.77E-01
Q_{40}	1.81E-02	4.45E-02	3.59E-01	2.87E-01	3.33E-01	4.66E-01
Q_{41c}	1.53E-02	1.36E-01	2.21E-01	1.64E-01	3.67E-01	2.92E-01
Q_{41s}	2.51E-02	5.81E-02	2.49E-01	2.40E-01	4.01E-01	3.11E-01
Q_{42c}	1.83E-02	5.63E-02	4.77E-01	4.93E-01	2.79E-01	3.07E-01
Q_{42s}	1.43E-02	7.18E-02	1.76E-01	4.01E-01	3.03E-01	3.72E-01
Q_{43c}	2.29E-02	1.40E-01	4.88E-01	3.43E-01	3.47E-01	3.43E-01
Q_{43s}	1.91E-02	6.59E-02	3.61E-01	5.03E-01	3.70E-01	3.92E-01
Q_{44c}	1.83E-02	1.92E-01	2.02E-01	7.29E-01	7.51E-01	4.78E-01
Q_{44s}	1.55E-02	5.63E-02	3.46E-01	3.35E-01	6.87E-01	4.15E-01