

Quaternion-based reorientation conditions for molecular dynamics analyses

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The need for a formal definition of a reorientation event in molecular dynamics simulations is recognised, and this is furnished through the quaternion formalism. Any rotation can be represented by a unit four-dimensional vector, and the general vector representing a molecule's orientation must be compared with those which represent symmetry operations. The vector dot-product is used to decide whether a given orientation is closer to the undisplaced or to the symmetry rotated orientation. When the latter occurs a reorientation event is recorded and the inverse symmetry operation is invoked.

Random reorientation rates are measured by a random walk procedure, and give a basis for an objective analysis of simulation results. The reorientation conditions are then extended by the introduction of adjustable parameters in order to change the random probabilities, resulting (for instance) in the ability to identify 2-fold tetrahedral events whereas the procedure first outlined cannot recognise these events.

All the relevant crystallographic rotation groups are considered, octahedral, tetrahedral, hexagonal, trigonal, tetragonal. For each of these the modified conditions are investigated.

1. INTRODUCTION

In the condensed state of molecular matter the thermal motion is mainly in the form of small displacements and rotations from mean positions. However, occasionally reorientational motion takes place and rotational diffusion is said to occur. This phenomenon can occur in a solid with no stacking defects which would otherwise facilitate translational diffusion. Usually reorientations are isolated events, but in some materials they can become very frequent especially above a transition temperature, whereupon the material is said to be in the plastic crystalline phase. Gaining an understanding of the reorientational characteristics of the molecules in this phase is an important aim of molecular dynamics simulations of these systems. The reorientational motion is not usually simply about one axis of the molecule, and it becomes important to be able to distinguish between reorientations about different axes. The quaternion formulation for molecular orientation is now generally accepted for molecular dynamics simulations [1] as it

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does not have the pathological behaviour of the Euler angle formulation, and so quaternions are used throughout this paper.

A molecular may reorient between (a) molecular symmetry related positions, (b) positions related by crystal symmetry but not molecular symmetry or (c) positions bearing no relationship with one another. For case (a) the molecular symmetry is often the crystal site symmetry, but is not always so; the site symmetry is either the same as that of the molecule or is a subgroup. For case (b) the symmetry of the molecule may contain a subgroup of the site symmetry, but this can only come about by the generation of crystal symmetry by existence of orientational disorder. In what follows we treat only the molecular symmetry reorientations, as this treatment contains all that is necessary for (b). No general analysis can be given for (c) as these cases require tests determined by the crystal structure of the particular system concerned.

The particular example which prompts this work is the body-centred cubic SF_6 system comprising octahedral molecules. Early work on this system [2] used a simple test for reorientation about the $\langle 100 \rangle$ axes only. In this paper we use the crystallographic notation for $\langle \ \rangle$ brackets, so that $\langle 100 \rangle$ means 'all rotations related to (100) by symmetry'. Subsequent work [3] suggested that $\langle 111 \rangle$ rotations are of some importance, and this raises two questions:

- (i) What is the definition of a reorientation?
- (ii) What is the probability of each possible reorientation if the rotational diffusion is entirely random?

In what follows we suggest an answer to (i) based on the quaternion formulation [1, 4] and a method for determining (ii), and give a number of results for various relevant symmetry groups.

To highlight the importance of the answers to (ii) the reader is challenged to make a judgement if presented with the following data. In a plastic crystalline system of octahedral SF_6 molecules 75 per cent of recorded reorientations were $\pi/2$ about $\langle 100 \rangle$ whereas 25 per cent were $2\pi/3$ about $\langle 111 \rangle$. Is this random behaviour, or is one of the reorientation processes dominant?

2. REORIENTATIONS USING QUATERNIONS

The general displacement of a molecule is, by Chasle's theorem, a translation of the centre of gravity of that molecule plus a rotation about an axis through an angle. The quaternion formulation contains the information about both the axis of rotation and the angle of rotation, thus allowing a molecule expressed in any coordinate system to be oriented in a molecular dynamics sample. In what follows it is assumed that all molecules are represented through a mean quaternion, and that we are dealing with *relative* reorientations away from this mean. Thus at some stage we need the unit quaternion, $\mathbf{1}$, representing no reorientation away from the mean.

The procedure to be adopted can be described as follows. At any one moment a molecule may be rotated away from the mean orientation through an angle α_i about some axis. However, because of symmetry the molecule could equally well be described as rotated about a different axis through α_j , and there will be a value of α for each possible molecular symmetry operation. In the course of a molecular dynamics calculation all the values of α will change smoothly so that, although we

choose to describe the molecular orientation through the smallest angle, α_i , in a reorientation event one of the values of α_j becomes the smallest. This then defines the event, and the present paper is concerned with determining the relative values of α which can be done most conveniently through the quaternion dot product.

The quaternion equations from Du Val [4] which are pertinent to this work are given in [2], where it is shown that if the orientational displacement of the molecule is represented by the quaternion

$$\mathbf{q} = (q_0; q_1, q_2, q_3) \quad (1)$$

and that this rotation approaches a molecular symmetry operation \mathbf{S} , then \mathbf{q} can be replaced by a new quaternion

$$\mathbf{q}' = \mathbf{q} * \mathbf{S}^{-1}, \quad (2)$$

and a reorientational event recorded. The asterisk here denotes quaternion multiplication—it is not a standard symbol but does add clarity in this paper. Postmultiplication by the inverse is required as the quaternion is used to get the molecule into the displaced orientation by acting on an undisplaced (mean) molecule, and any molecular symmetry operation on the undisplaced molecule will not alter the disposition of the final molecule.

In [2] it is shown that a quaternion approaching a positive $\pi/2$ reorientation about (001) can be replaced by

$$\mathbf{q}' = \mathbf{q} * \left(\frac{1}{\sqrt{2}}; 0, 0, -\frac{1}{\sqrt{2}} \right). \quad (3)$$

For clarity the coefficient for the real axis is separated from the three coefficients for the 'imaginary prime' [4] by a semi-colon, though it should be remembered that the four coefficients all behave alike in that they form a unit vector in a four-dimensional Euclidean space. In this representation the unit quaternion

$$\mathbf{1} = (1; 0, 0, 0) \quad (4)$$

corresponds to the undisplaced mean molecule, and we now ask whether \mathbf{q} or \mathbf{q}' of equation (3) is closer to $\mathbf{1}$, and then if \mathbf{q}' is the closer it is taken as the new quaternion and a reorientation is recorded. Performing the quaternion multiplication we get

$$\mathbf{q}' = \left(\frac{q_3 + q_0}{\sqrt{2}}; \frac{q_1 - q_2}{\sqrt{2}}, \frac{q_2 + q_1}{\sqrt{2}}, \frac{q_3 - q_0}{\sqrt{2}} \right). \quad (5)$$

The test that can be applied is the comparison of the (four-dimensional) dot-product of \mathbf{q} and the unit quaternion $\mathbf{1}$ with a dot-product of \mathbf{q}' also with the unit quaternion $\mathbf{1}$. These are respectively q_0 and $(q_3 + q_0)/\sqrt{2}$. Reorientation is recorded if

$$\frac{q_3 + q_0}{\sqrt{2}} > q_0 \quad (\text{i.e. } q_3 > 0.414q_0). \quad (6)$$

As there are six possible four-fold reorientations which we will denote as $\pi/2 \langle 100 \rangle$, the condition (6) is generalised with q_3 replaced by the largest magnitude of q_1, q_2 or q_3 say $|q_{\max}|$,

$$|q_{\max}| > 0.414q_0. \quad (7)$$

In condition (7) and the other conditions later it is assumed that q_0 is always positive. This choice can always be made; the quaternion representation of orientations is double valued in such a way that \mathbf{q} and $-\mathbf{q} = (-q_0, -q_1, -q_2, -q_3)$ give the same orientation.

There are two other classes of symmetry operation that must be considered for an octahedral molecule, the $2\pi/3 \langle 111 \rangle$ and the $\pi \langle 110 \rangle$. If the quaternion approaches $2\pi/3 \langle 111 \rangle$, then

$$\begin{aligned} \mathbf{q}' &= \mathbf{q} * \left(\frac{1}{2}; -\frac{1}{2}, -\frac{1}{2}, -\frac{1}{2} \right) \\ &= \left(\frac{q_1 + q_2 + q_3 + q_0}{2}; \frac{q_1 - q_2 + q_3 - q_0}{2}, \right. \\ &\quad \left. \frac{q_1 + q_2 - q_3 - q_0}{2}, \frac{-q_1 + q_2 + q_3 - q_0}{2} \right) \end{aligned} \quad (8)$$

giving reorientation if

$$\frac{q_1 + q_2 + q_3 + q_0}{2} > q_0. \quad (9)$$

It is possible for such a condition to be reached before (7) and therefore for such a reorientation to be recorded. This can be seen by taking the most favourable set of quaternion coefficients $q_1 = q_2 = q_3$ which makes the condition

$$\frac{3q_3 + q_0}{2} > q_0 \quad (\text{i.e. } q_3 > 0.333q_0), \quad (10)$$

which is then compared with (7).

Generalisation of (9) to include all the eight possible $2\pi/3 \langle 111 \rangle$ reorientations is easily found to be

$$\frac{|q_1| + |q_2| + |q_3| + q_0}{2} > q_0 \quad (11)$$

Consider now the search for a reorientation about the diad axis of the octahedron along $\langle 110 \rangle$. This symmetry operation is

$$\mathbf{S}_d = \left(0; \frac{1}{\sqrt{2}}, \frac{1}{\sqrt{2}}, 0 \right) \quad (12)$$

for which the new quaternion value \mathbf{q}' is

$$\mathbf{q}' = \mathbf{q} * \mathbf{S}_d^{-1} = \left(\frac{q_1 + q_2}{\sqrt{2}}; \frac{q_3 - q_0}{\sqrt{2}}, \frac{-q_3 - q_0}{\sqrt{2}}, \frac{q_2 - q_1}{\sqrt{2}} \right). \quad (13)$$

The reorientation condition is now

$$\frac{q_1 + q_2}{\sqrt{2}} > q_0. \quad (14)$$

It can now be shown that such a reorientation of the octahedron will never be counted because a $\pi/2 \langle 100 \rangle$ reorientation will be recorded before (14) can happen. The most favourable coefficients for the diad operation on \mathbf{q} have

$$q_1 = q_2 \quad \text{giving} \quad q_1 > 0.707q_0. \quad (15)$$

Clearly condition (7) must occur before (15) is reached and thus there is no possible classification for a $\pi \langle 110 \rangle$ reorientation. This result is confirmed by the results of the next section.

3. RANDOM REORIENTATION PROBABILITIES

Consider an octahedral molecule starting in an undisplaced orientation undergoing a succession of small random orientational changes such that when conditions (7) or (11) are met the appropriate quaternion change is applied and a reorientation is counted. For true random motion there will be a fixed ratio between the frequencies of the two possible reorientation classes. This ratio must be known before the systematics of the motion in any system can be properly analysed.

A quaternion is represented by a point on the unit four-dimensional hypersphere. A certain volume on the hypersphere corresponds to reorientations closest to the unit quaternion (no reorientation), and there are surfaces separating this volume and those volumes associated with reorientations which are closest to the various point-group symmetry operations. A small reorientation change is represented by an arc of proportional length on the hypersphere, and this proportion is constant over the whole hypersphere. Therefore the reorientational frequencies we need are proportional to the surface areas on the hypersphere which separate the volumes just mentioned. The hypersphere divisions are shown schematically in figure 1. The problem must have an analytic solution through the use of four-dimensional spherical trigonometry but we use a simpler method here.

Instead of developing the analytic result it is sufficient for our purposes to solve the problem by a random walk procedure. Such a possibility is evident from the paragraph above. Starting at an undisplaced orientation, small orientation changes are made at random using a pseudo-random number generator to choose the step, and the frequency of reorientations is found by running the calculation for a sufficient time and performing the reorientation checks outlined in the previous section. As the quaternion checks required for the random walk are exactly as needed in the MD work on a simulated system the random walk method is the natural way of determining the random rate frequencies and gives rise to tested computer software for the MD work. Tests of the software include

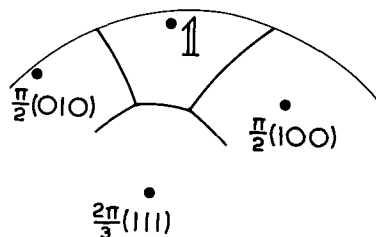


Figure 1. Schematic representation of the 3-*d* 'surface' of the 4-*d* unit hypersphere divided by octahedral symmetry. The point 1 corresponds to no rotation, and the three other points depicted represent four-fold rotations about (100) and (010) and a three-fold rotation about (111). The lines depict areas separating the volumes of the hypersphere surface which are nearest to the rotation points according to the dot-product rule.

the requirement that the reorientation rate frequency about each of the eight $\langle 111 \rangle$ axes should be the same, likewise for the six $\langle 100 \rangle$ reorientations. The variations between the separate estimates of each rate can then be used to give estimates of the accuracy of the MC determination of the rate frequencies. These are the errors quoted with the results below, and serve to indicate the number of independent events used in the calculation.

The calculations were all implemented on the ICL Distributed Array Processor (DAP) in Edinburgh [5]. Each of the 4096 processing elements performs a separate random walk calculation, and the computer runs at its maximum 25 Mflops capacity throughout. Accuracy of about 1 per cent is generally achieved in about 20 s of running time.

In the analysis of the motion of octahedral molecules, condition (15) was included and no such event was recorded. This was used as another software consistency check. The resulting rates for $\pi/2 \langle 100 \rangle$ and $2\pi/3 \langle 111 \rangle$ were found to be 89.2 and 10.8 per cent (± 0.1), and therefore the answer to the question posed in the introduction is that the 25 per cent $\langle 111 \rangle$ reorientation is far more frequent than expected for random motion—such a conclusion may not have been anticipated!

4. OTHER SYMMETRIES

The symmetry groups of interest are the proper rotation point groups which could include the icosahedral group, but this is left for the ambitious reader. Only the crystallographic groups involving more than one class of symmetry operation besides the identity are considered here.

(i) Octahedral group (e.g. SF_6)

(i, a) condition (7)

(i, b) condition (11)

(i, c) condition (14) generalized.

Random rates (i, a) 89.2 per cent, (i, b) 10.8 per cent, (i, c) 0 per cent.

The rate for (i, a) is appropriate for the motion of a tetrahedral molecule between two sites which, on average, are related by the crystal site symmetry. Rates (i, b) and (i, c) apply for the tetrahedral molecule as the symmetry operations are in the tetrahedral subgroup.

(ii) Tetrahedral group (e.g. CBr_4)

(ii, a) $2\pi/3 \langle 111 \rangle$ as for (i, b)

(ii, b) $\pi \langle 001 \rangle$ $\mathbf{S}^{-1} = (0; 0, 0, -1)$, (particular example)

$\mathbf{q}' = \mathbf{q} * \mathbf{S}^{-1} = (q_3; -q_2, q_1, -q_0)$

Reorient if $q_3 > q_0$ (q_3 positive)

Again we find that for the most favourable \mathbf{q} (i.e. $q_1 = q_2 = 0$) the reorientation condition for (ii, a) becomes

$$\frac{|q_3| + q_0}{2} > q_0 \quad (\text{i.e. } |q_3| > q_0)$$

Thus (ii, a) will occur before (ii, b).

(iii) *Hexagonal, 62 (e.g. benzene)*

$$(iii, a) \frac{2\pi}{6} (0 \ 0 \ \pm 1) \quad \mathbf{S}^{-1} = \left(\frac{\sqrt{3}}{2}; 0, 0, \frac{1}{2} \right)$$

Reorient if $\frac{|q_3| + \sqrt{3}q_0}{2} > q_0$ (i.e. $|q_3| > (2 - \sqrt{3})q_0$)

$$(iii, b) \frac{2\pi}{2} (\pm 1 \ 0 \ 0) \left[\& \frac{2\pi}{2} \left(\pm \frac{1}{2} \pm \frac{\sqrt{3}}{2} \ 0 \right) \right] \quad \mathbf{S}^{-1} = (0; \mp 1, 0, 0)$$

Reorient if $|q_1| > q_0$ [& similar conditions]

No conflict between (iii, a) and (iii, b).

Random rates (iii, a) 76.5 per cent, (iii, b) 23.5 per cent.

(iv) *Trigonal, 32 (e.g. s-triazine)*

$$(iv, a) \frac{2\pi}{3} (0 \ 0 \ \pm 1) \quad \mathbf{S}^{-1} = \left(\frac{1}{2}; 0, 0, \mp \frac{\sqrt{3}}{2} \right)$$

Reorient if $\frac{\sqrt{3}|q_3| + q_0}{2} > q_0$ (i.e. $\sqrt{3}|q_3| > q_0$)

(iv, b) Diads similar to example in (iii, b).

No conflict between (iv, a) and (iv, b).

Random rates (iv, a) 51.2 per cent, (iv, b) 48.8 per cent

(v) *Tetragonal, 42 (e.g. octasulphur)*

$$(v, a) \frac{2\pi}{4} (0 \ 0 \ \pm 1) \quad \text{As (i, a)}$$

(v, b) Diads similar to example in (iii, b).

No conflict between (v, a) and (v, b).

Random rates (v, a) 65.2 per cent, (v, b) 34.8 per cent.

5. MORE GENERAL CONDITION

(i) *Octahedral*

The conditions for claiming a reorientation as presented so far appear to be too strict in a number of cases. Take for instance the case where an octahedral molecule is rotating steadily about (110). In the formulation above no (110) reorientation can be recorded, but after a partial reorientation condition (7) will be met in some form. The molecule is then deemed to have reoriented about (say) (100), but the new quaternions immediately satisfy a (111) reorientation. After a further rotation about this second axis the condition for a (111) rotation is met. This succession of three rapid reorientations is equivalent to a single (110) reorientation, and it is therefore possible in principle to identify all possible $\langle 110 \rangle$ reorientations from such event sequences. Such a procedure is very inconvenient, and raises the very sort of question we are trying to avoid with the quaternion test procedure, namely: when does one accept such a triple sequence of $\langle 100 \rangle$ and $\langle 111 \rangle$ events as a single $\langle 110 \rangle$ event? A procedure must be sought which allows $\langle 110 \rangle$ events (and events (ii, b)) to be identified in their own right, and although such a procedure may require arbitrary parameters the important result to know is what the random rate would be for the chosen parameter values.

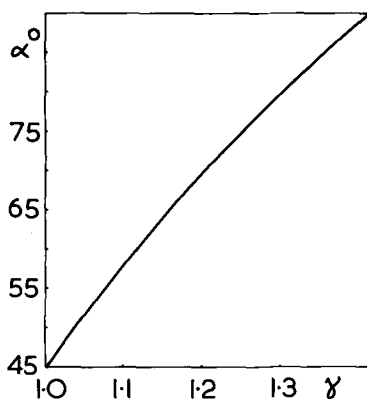


Figure 2. Variation of angle required for recording a $(\pi/2)$ $\langle 100 \rangle$ reorientation, α , as a function of γ . When $\gamma = 1$ the reorientation is recorded for $\alpha = 45^\circ$, this being half-way to the $\pi/2$ orientation.

Condition (7) is a comparison of $\mathbf{q} \cdot \mathbf{l}$ and $\mathbf{q}' \cdot \mathbf{l}$, requiring $\mathbf{q}' \cdot \mathbf{l}$ to be the larger before implementing reorientation. We now suggest that

$$\mathbf{q}' \cdot \mathbf{l} > \gamma \mathbf{q} \cdot \mathbf{l}, \quad (16)$$

where $\gamma (> 1)$. For the $\pi/2$ $\langle 100 \rangle$ reorientation of the octahedron we get

$$\frac{q_1 + q_0}{\sqrt{2}} > \gamma q_0$$

and for the ideal case ($q_2 = q_3 = 0$) we find that reorientation is accepted when the rotation has reached the angle α as shown in figure 2. Thus we see that for $\gamma = \sqrt{2}$ the rotation must be the complete $\pi/2$ before acceptance, and so such a value of γ is probably rather too large for practical use. For any value below $\gamma = \sqrt{2}$ the $\pi/2$ $\langle 100 \rangle$ reorientation is always recorded in preference to the π $\langle 110 \rangle$, and so there is no way in which the introduction of a single parameter γ allows these latter reorientations to be recorded and this simple expedient fails.

One advantage of using γ for the octahedral case is that the rate ratios change as shown in figure 3. In the example of the question posed to the reader the $(2\pi/3)$

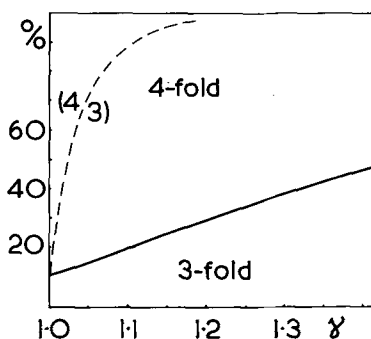


Figure 3. Variation of the rate ratio for four-fold and three-fold reorientations as a function of γ , ($\gamma_2 = \gamma_3 = \gamma_4$). Here we observe that the three-fold reorientation rate increases from 10.8 per cent at $\gamma = 1$, reaching 40 per cent at $\gamma \sim 1.32$. The dashed curve is for $\gamma_2 = \gamma_3 = 1$, $\gamma_4 = \gamma$, labelled in parentheses, showing a much more rapid three-fold rate increase as a function of γ_4 .

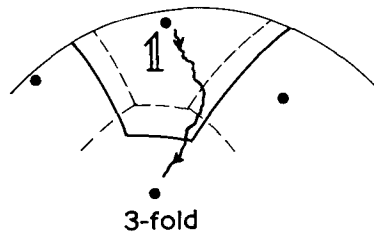


Figure 4. Schematic representation of the new hypersphere divisions (see figure 1) for octahedral symmetry with $\gamma \sim 1.2$. The random walk path shown is such that a $(\pi/2)$ (100) reorientation would be recorded if $\gamma = 1$.

$\langle 111 \rangle$ reorientations were significantly greater than by chance, and the choice of γ (say $\gamma = 1.2$) makes the recording of the genuine events more reliable, reducing the number of artificial $(\pi/2)$ $\langle 001 \rangle$ events. Figure 4 shows schematically the effect of introducing γ into the condition for octahedral reorientations. The boundaries of figure 1 have been shifted away from the **1** position, thus changing their relative volumes. A possible actual path on the hypersphere for a $(2\pi/3)$ (111) reorientation is shown by the arrowed line. With the strict $\gamma = 1$ condition a $(\pi/2)$ (100) reorientation would be first recorded, whereas with γ increased the excursion towards $(\pi/2)$ (100) has no effect and only the $(2\pi/3)$ (111) is recorded.

It turns out to be possible to modify the condition (16) further and achieve a situation where $\pi \langle 110 \rangle$ events are recorded. For this the condition

$$\mathbf{q}' \cdot \mathbf{1} > \gamma_S \mathbf{q} \cdot \mathbf{1} \quad (17)$$

where different values of γ are introduced for the different symmetry operations. The result shown in figure 3 has been obtained by increasing γ_2 , γ_3 and γ_4 (the two-fold, three-fold and four-fold symmetry parameters) equally. The dashed curve on this figure shows the result where $\gamma_2 = \gamma_3 = 1$ and only γ_4 is varied. Figure 5 shows the result for $\gamma_2 = 1$ and with $\gamma_3 = \gamma_4$ increasing, where 2-fold reorientations eventually become detectable. From the reorientation conditions it can be shown that no $\langle 110 \rangle$ event can occur until both γ_3 and γ_4 exceed $(1 + \sqrt{2})/2$. Figure 2 shows that for γ_4 this special value requires 70° rotation about the four-fold axis before reorientation is accepted. Such large values of γ_3 and γ_4 could be used in those cases where $\langle 110 \rangle$ reorientation is expected to be impor-

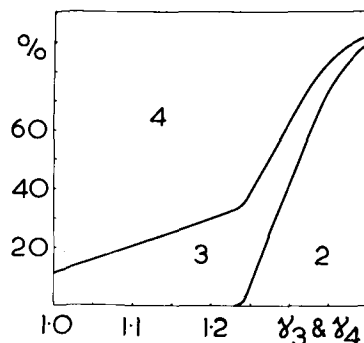


Figure 5. Rate ratios for octahedral symmetry as a function of $\gamma_3 = \gamma_4$, with $\gamma_2 = 1$. two-fold reorientations (denoted 2) begin to be possible when γ_3 and γ_4 exceed $(1 + \sqrt{2})/2$.

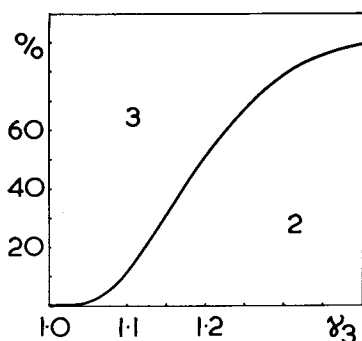


Figure 6. Rate ratios for tetrahedral symmetry as a function of γ_3 , with $\gamma_2 = 1$. two-fold and three-fold reorientations (denoted 2 and 3) become nearly equally probable at $\gamma_3 = 1.2$.

tant, whereas if it is only the $\langle 111 \rangle$ reorientations which need to be emphasized then we could keep $\gamma_2 = \gamma_3 = 1$ and choose a value for γ_4 from figure 3 (broken line).

The use of γ values greater than unity has one clear advantage. If a molecule spends a period of time half-way between two symmetry sites it could cross the $\gamma = 1$ border many times without undergoing large displacements, and each cross-

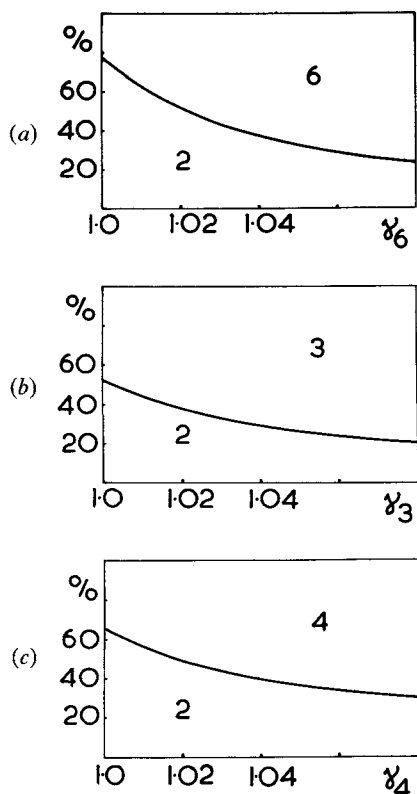


Figure 7. Rate ratios for (a) hexagonal, (b) trigonal and (c) tetragonal symmetries as functions of γ_6 , γ_3 and γ_4 respectively, with $\gamma_2 = 1$.

ing should not be regarded as a reorientation. For this reason the current values being used for the analysis of SF₆ results is line (b) of the table which follows

Random rates for motion in an octahedral site for $\gamma_2 = 1$.

	γ_3	γ_4	Random rates (per cent)	
			Three-fold	Four-fold
(a)	1.02	1.06	43.5	56.5
(b)	1.03	1.09	52.1	47.9
(c)	1.04	1.12	59.5	40.5

(ii) Tetragonal

The condition (ii, b) becomes immediately possible if (ii, a) is modified by any $\gamma_3 > 1$. The rate probabilities are shown in figure 6 as a function of γ_3 , with equal probabilities occurring at $\gamma_3 = 1.195(5)$.

(iii-v) Uniaxial groups

Although there is no proper reorientation event which could not be recorded with the conditions for the uniaxial n -fold groups, we present for completeness in figure 7 the change in rate ratios as a function of γ_n . This choice accentuates the recording of two-fold events, as these are the least likely events in actual systems of molecules of these symmetries.

6. CONCLUSIONS

A formal method has been presented for analysing molecular dynamics calculations for molecular reorientations. Without a formal procedure conclusions concerning the most probable reorientation are subjective and could lead to a wrong interpretation of computer simulations. The need for this formalism arose from the analysis of the plastic hase simulation of SF₆, where many cases were observed in which multiple reorientation events seemed to occur with an unphysical rapidity. Much of this work, which follows on from that of Dove and Pawley [3], is being repeated using the conditions outlined herein.

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