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ORGANOMETALLIC COMPOUNDS
XVII*. MATRIX REPRESENTATION OF OCTAHEDRAL COMPLEXES AND OF THEIR INTRAMOLECULAR ISOMERIZATIONS VIA TRIGONAL OR RHOMBIC TWISTS

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SUMMARY

Matrix representations are given for six-coordinate structures (product matrices *P*) and for their intramolecular isomerization reactions via trigonal or rhombic twists (reaction matrices *R*). Matrix transformations are also described, which allow to deduce from the general case the subset of rearrangements generated by various constraints. Several examples are depicted and are compared with previous descriptions.

INTRODUCTION

Besides topological[2,3] and non-topological[4] representations of intramolecular isomerizations via pseudo-rotations of trigonal-bipyramidal structures, matrix representations have also been described[1].

Topological[5] and non-topological[6] descriptions may be given for the analogous rearrangements via trigonal or rhombic twists of octahedral complexes. It seemed interesting to see whether the matrix representation used to represent the processes in five-coordinate species could be extended to the present problem.

RESULTS

Matrix representation of octahedral structures

A set of three matrices (*1P*, *2P* and *3P*) is needed to represent the 30 octahedral isomers (a to q and $\bar{a}$ to $\bar{q}$) of a six-coordinate complex 123,456*. This difference between six- and five-coordinate complexes arises from the fact that, in every octahedral structure, *three* couples of ligands are in *trans* position, when only *two* substituents are *trans* with respect to each other (AA) in trigonal-bipyramidal molecules.

These three matrices thus relate, as in the case of five-coordinate complexes[1], the relative *trans* position of the ligands with the name (symbol) of the different isomers.

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* For Part XVI see ref. 1.

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If one applies the numbering* which has been defined in ref. 6 and if one uses letters instead of digits to describe the different isomers, which avoids the confusion between ligands and isomers[3], one gets

$$\begin{array}{c|cccccc} & 1 & 2 & 3 & 4 & 5 & 6 \\ \hline 1 & 0 & \bar{i} & \bar{l} & \bar{m} & a & q \\ 2 & i & 0 & d & \bar{c} & \bar{q} & \bar{a} \\ 3 & l & \bar{d} & 0 & q & c & n \\ 4 & m & c & \bar{q} & 0 & \bar{d} & j \\ 5 & \bar{a} & q & \bar{c} & d & 0 & g \\ 6 & \bar{q} & a & \bar{n} & \bar{j} & \bar{g} & 0 \end{array} = \mathit{1P}; \quad \begin{array}{cccccc} 1 & 2 & 3 & 4 & 5 & 6 \\ \hline 0 & k & \bar{h} & g & j & \bar{d} \\ \bar{k} & 0 & \bar{g} & h & e & l \\ h & g & 0 & \bar{k} & f & i \\ \bar{g} & \bar{h} & k & 0 & \bar{i} & \bar{f} \\ \bar{j} & \bar{e} & \bar{f} & i & 0 & k \\ d & \bar{l} & \bar{i} & f & \bar{k} & 0 \end{array} = \mathit{2P}; \quad \begin{array}{cccccc} 1 & 2 & 3 & 4 & 5 & 6 \\ \hline 0 & \bar{f} & \bar{e} & b & n & c \\ f & 0 & \bar{j} & \bar{n} & \bar{b} & m \\ e & j & 0 & a & m & \bar{b} \\ \bar{b} & n & \bar{a} & 0 & l & e \\ \bar{n} & b & \bar{m} & \bar{l} & 0 & h \\ \bar{c} & \bar{m} & b & \bar{e} & \bar{h} & 0 \end{array} = \mathit{3P}$$

Matrix transformations

The matrices *1P*, *2P* and *3P* used simultaneously define all the structures which have two ligands k and l in *trans* position ($1p_{kl}$, $1p_{lk}$, $2p_{kl}$, $2p_{lk}$, $3p_{kl}$ and $3p_{lk}$, p_{lk} being always the optical antipode of p_{kl}).

Let us define a matrix $C(\mathrm{kl}, t)$** which is a p by p zero matrix where c_{kk} and c_{ll} have been replaced by 1 (ref. 1); the operation

$$\tilde{C}(\mathrm{kl}, t) \times P \times C(\mathrm{kl}, t) = P(\mathrm{kl}, t)^{***}$$

changes each of the original P matrices into the particular ones $P(\mathrm{kl}, t)$ which describe the case where ligands k and l are in *trans* position

When kl is a chelating ligand†, k and l cannot be *trans* with respect to one another and one finds

$$P(\mathrm{kl}, \bar{t}) = P - P(\mathrm{kl}, t)$$

These matrix transformations have to be performed with each of the three matrices *1P*, *2P* and *3P* to get all the excluded isomers.

The changes which occur when two ligands k and l are identical are no more caused by structures which are impossible to form, but by isomers which become identical to one another. The restrictions may be seen on the matrix obtained from P by setting k = l: the three six by six matrices P may then be reduced to three five by five ones $P(\mathrm{k=l})$. $p_{kx} = p_{lx}$ and $p_{xk} = p_{xl}$ (if $x \neq k$, l) and $p_{kl} = p_{lk}$ express the identity of isomers. This may also be represented by a matrix transformation[1]

$$\tilde{C}(\mathrm{k=l}) \times P \times C(\mathrm{k=l}) = P(\mathrm{k=l})$$

where $C(\mathrm{k=l})$ is a rectangular matrix constructed from a (p − 1) by (p − 1) unit matrix by inserting between lines (l − 1) and l, a row which is identical to line k††.

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* For the numbering convention and symbolism see ref. 6; a = 123,456; b = 123,645; c = 123,564; d = 125,463; e = 126,435; f = 136,425; g = 125,643; h = 126,534; i = 135,426; j = 124,653; k = 135,624; l = 125,436; m = 125,346; n = 123,654 and q = 123,465.

** The C matrices describe the reduction or original matrices under the influence of constraints; the type of constraint is represented in parentheses.

*** $\tilde{C}(\mathrm{kl}, t)$ is the transposed of $C(\mathrm{kl}, t)$; in this particular case, $\tilde{C}(\mathrm{kl}, t) = C(\mathrm{kl}, t)$.

† The case of complexes with two or three bidentate ligands will be examined elsewhere[5].

†† Some of the restrictions (x = y) are obtained in duplicate.

Matrix representations of intramolecular rearrangements via trigonal or rhombic twists of octahedral structures

The intramolecular isomerization reactions of $ML_{6'}$, described in refs. 4 and 5, may be represented by a square 30 by 30 symmetrical rearrangement matrix R, whose coefficients r_{yz} are equal to 1 if isomer y may be converted into isomer z by one trigonal or rhombic twist, and 0 otherwise

$$R = \left(\begin{array}{c|c} A & B \\ \hline \tilde{B} & \tilde{A} \end{array}\right)$$

$$\begin{array}{c|ccccccccccccccc|}
 & a & b & c & d & e & f & g & h & i & j & k & l & m & n & q \\
a & 0&1&1&1&1&1&1&1&1&0&0&0&0&0&0 \\
b & 1&0&1&0&0&0&0&0&1&1&1&0&0&0&0 \\
c & 1&1&0&0&0&0&0&0&0&0&0&0&0&0&0 \\
d & 1&0&0&0&1&0&0&0&0&0&0&0&1&1&0 \\
e & 1&0&0&1&0&0&0&0&0&0&1&0&0&0&0 \\
f & 1&0&0&0&0&0&1&0&0&0&0&0&0&0&1 \\
g & 1&0&0&0&0&1&0&0&0&0&0&1&0&1&0 \\
h & 1&0&0&0&0&0&0&0&1&0&0&0&0&0&0 \\
i & 1&0&0&0&0&0&0&1&0&1&0&0&1&0&1 \\
j & 0&1&0&0&0&0&0&0&1&0&0&0&0&0&0 \\
k & 0&1&0&0&1&0&0&0&0&0&0&0&1&1&0 \\
l & 0&1&0&0&0&0&1&0&0&0&0&0&0&0&1 \\
m & 0&0&0&1&0&0&0&0&1&0&1&0&0&0&0 \\
n & 0&0&0&1&0&0&1&0&0&0&1&0&0&0&1 \\
q & 0&0&0&0&0&1&0&0&1&0&0&1&0&1&0
\end{array} = A; \quad
\begin{array}{c|ccccccccccccccc|}
 & \bar{a} & \bar{b} & \bar{c} & \bar{d} & \bar{e} & \bar{f} & \bar{g} & \bar{h} & \bar{i} & \bar{j} & \bar{k} & \bar{l} & \bar{m} & \bar{n} & \bar{q} \\
a & 0&0&0&0&0&0&0&0&0&0&0&0&0&0&0 \\
b & 0&0&0&1&0&1&0&1&0&0&0&0&0&0&0 \\
c & 0&0&0&0&1&0&1&0&1&1&1&1&0&0&0 \\
d & 0&1&0&0&0&1&0&1&0&0&1&0&0&0&0 \\
e & 0&0&1&0&0&0&1&0&1&0&0&0&1&1&0 \\
f & 0&1&0&1&0&0&0&1&0&0&0&1&0&1&0 \\
g & 0&0&1&0&1&0&0&0&1&0&0&0&0&0&1 \\
h & 0&1&0&1&0&1&0&0&0&1&0&0&1&0&1 \\
i & 0&0&1&0&1&0&1&0&0&0&0&0&0&0&0 \\
j & 0&0&1&0&0&0&0&1&0&0&1&1&1&0&1 \\
k & 0&0&1&1&0&0&0&0&0&1&0&1&0&0&0 \\
l & 0&0&1&0&0&1&0&0&0&1&1&0&0&1&0 \\
m & 0&0&0&0&1&0&0&1&0&1&0&0&0&1&1 \\
n & 0&0&0&0&1&1&0&0&0&0&0&1&1&0&0 \\
q & 0&0&0&0&0&0&1&1&0&1&0&0&1&0&0
\end{array} = B$$

Matrix transformations

It is also possible to transform the r by r matrix R representing the possible isomerization reactions of 123,456, into other smaller matrices which are valid for simpler structures.

The comparison of matrices P valid for a given system with matrices P valid for the general case ML_6, gives immediately the structures which are excluded and those which have become identical to other ones.

If structure p_{kl} is excluded, one may describe a matrix transformation which changes R into $R(p_{kl}=0)$, as for the five-coordinate complexes[1]. A rectangular transformation matrix $C(p_{kl}=0)$ has to be used, which can be constructed from a $(r-1)$ by $(r-1)$ unit matrix by inserting a row of zero's between line $(p_{kl}-1)$ and line p_{kl}. One has

$$\tilde{C}(p_{kl}=0) \times R \times C(p_{kl}=0) = R(p_{kl}=0)$$

If two structures m and n become equivalent, one has, as in the case of trigonal-bipyramidal structures[1]:

$$\tilde{C}(m=n) \times R \times C(m=n) = R(m=n)$$

where $C(m=n)$ is a rectangular matrix constructed from a $(r-1)$ by $(r-1)$ unit matrix by inserting between lines (n−1) and n, a row which is identical to line m (vide supra).

With the aid of these matrices *1P*, *2P*, *3P*, $C(\mathrm{kl}, t)$, $C(\mathrm{u}=\mathrm{w})$ and $C(\mathrm{z}=0)$, one can easily deduce the subset of isomerizations generated by various constraints which forbid the existence of certain isomers or which render two isomers of the general case identical to one another. Several examples will now be examined and compared with topological representations which have already been described before.

Example 1 : 123,455. The general matrix *1P* may be transformed into *1P*(5=6) according to

$$\begin{bmatrix} 1&0&0&0&0&0 \\ 0&1&0&0&0&0 \\ 0&0&1&0&0&0 \\ 0&0&0&1&0&0 \\ 0&0&0&0&1&1 \end{bmatrix} \times \mathit{1P} \times \begin{bmatrix} 1&0&0&0&0 \\ 0&1&0&0&0 \\ 0&0&1&0&0 \\ 0&0&0&1&0 \\ 0&0&0&0&1 \\ 0&0&0&0&1 \end{bmatrix} = \begin{array}{c|ccccc} & 1 & 2 & 3 & 4 & 5=6 \\ \hline 1 & 0 & \bar{i} & \bar{l} & \bar{m} & a+q \\ 2 & i & 0 & d & \bar{c} & \bar{q}+\bar{a} \\ 3 & l & \bar{d} & 0 & q & c+n \\ 4 & m & c & q & 0 & \bar{d}+j \\ 5=6 & \bar{a}+\bar{q} & a+q & \bar{c}+\bar{n} & d+\bar{j} & g+\bar{g} \end{array}$$

The same may be done for *2P* and *3P*. One gets finally $a=q$, $\bar{a}=\bar{q}$, $b=\bar{m}$, $\bar{b}=m$, $c=n$, $\bar{c}=\bar{n}$, $d=\bar{j}$, $\bar{d}=j$, $e=l$, $\bar{e}=\bar{l}$, $f=i$, $\bar{f}=\bar{i}$, $g=\bar{g}$, $h=\bar{h}$, $k=\bar{k}$.

It is now possible to determine $R(5=6)=R(123{,}455)$, using the suitable transformation matrix defined from $P(5=6)$. The matrix $C(a=q, \bar{a}=\bar{q}, b=\bar{m}, \bar{b}=m, c=n, \bar{c}=\bar{n}, d=\bar{j}, \bar{d}=j, e=l, \bar{e}=\bar{l}, f=i, \bar{f}=\bar{i}, g=\bar{g}, h=\bar{h}, k=\bar{k})$ may thus be written

$$\begin{array}{c|cccccccccccccccccccccccccccccc} & a & b & c & d & e & f & g & h & i & j & k & l & m & n & q & \bar{a} & \bar{b} & \bar{c} & \bar{d} & \bar{e} & \bar{f} & \bar{g} & \bar{h} & \bar{i} & \bar{j} & \bar{k} & \bar{l} & \bar{m} & \bar{n} & \bar{q} \\ \hline a & 1&0&0&0&0&0&0&0&0&0&0&0&0&0&1&0&0&0&0&0&0&0&0&0&0&0&0&0&0&0 \\ b & 0&1&0&0&0&0&0&0&0&0&0&0&0&0&0&0&0&0&0&0&0&0&0&0&0&0&0&1&0&0 \\ c & 0&0&1&0&0&0&0&0&0&0&0&0&0&1&0&0&0&0&0&0&0&0&0&0&0&0&0&0&0&0 \\ d & 0&0&0&1&0&0&0&0&0&0&0&0&0&0&0&0&0&0&0&0&0&0&0&0&1&0&0&0&0&0 \\ e & 0&0&0&0&1&0&0&0&0&0&0&1&0&0&0&0&0&0&0&0&0&0&0&0&0&0&0&0&0&0 \\ f & 0&0&0&0&0&1&0&0&1&0&0&0&0&0&0&0&0&0&0&0&0&0&0&0&0&0&0&0&0&0 \\ g & 0&0&0&0&0&0&1&0&0&0&0&0&0&0&0&0&0&0&0&0&0&1&0&0&0&0&0&0&0&0 \\ h & 0&0&0&0&0&0&0&1&0&0&0&0&0&0&0&0&0&0&0&0&0&0&1&0&0&0&0&0&0&0 \\ k & 0&0&0&0&0&0&0&0&0&0&1&0&0&0&0&0&0&0&0&0&0&0&0&0&0&1&0&0&0&0 \\ \bar{a} & 0&0&0&0&0&0&0&0&0&0&0&0&0&0&0&1&0&0&0&0&0&0&0&0&0&0&0&0&0&1 \\ \bar{b} & 0&0&0&0&0&0&0&0&0&0&0&0&1&0&0&0&1&0&0&0&0&0&0&0&0&0&0&0&0&0 \\ \bar{c} & 0&0&0&0&0&0&0&0&0&0&0&0&0&0&0&0&0&1&0&0&0&0&0&0&0&0&0&0&1&0 \\ \bar{d} & 0&0&0&0&0&0&0&0&0&1&0&0&0&0&0&0&0&0&1&0&0&0&0&0&0&0&0&0&0&0 \\ \bar{e} & 0&0&0&0&0&0&0&0&0&0&0&0&0&0&0&0&0&0&0&1&0&0&0&0&0&0&1&0&0&0 \\ \bar{f} & 0&0&0&0&0&0&0&0&0&0&0&0&0&0&0&0&0&0&0&0&1&0&0&1&0&0&0&0&0&0 \end{array}$$

$$= \tilde{C} \begin{bmatrix} a=q,\ b=\bar{m},\ c=n,\ d=\bar{j},\ e=l,\ f=i, & g=\bar{g} \\ \bar{a}=\bar{q},\ \bar{b}=m,\ \bar{c}=\bar{n},\ \bar{d}=j,\ \bar{e}=\bar{l},\ \bar{f}=\bar{i}, & h=\bar{h} \\ & k=\bar{k} \end{bmatrix}$$

$$\begin{array}{c|ccccccccccccccc|}
 & a & b & c & d & e & f & g & h & k & \bar{a} & \bar{b} & \bar{c} & \bar{d} & \bar{e} & \bar{f} \\
\hline
a & 0 & 1 & 1 & 1 & 1 & 2 & 1 & 1 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
b & 1 & 0 & 1 & 0 & 1 & 0 & 0 & 1 & 1 & 0 & 0 & 0 & 2 & 0 & 1 \\
c & 1 & 1 & 0 & 1 & 0 & 0 & 1 & 0 & 1 & 0 & 0 & 0 & 0 & 2 & 1 \\
d & 1 & 0 & 1 & 0 & 1 & 0 & 0 & 1 & 1 & 0 & 2 & 0 & 0 & 0 & 1 \\
e & 1 & 1 & 0 & 1 & 0 & 0 & 1 & 0 & 1 & 0 & 0 & 2 & 0 & 0 & 1 \\
f & 2 & 0 & 0 & 0 & 0 & 0 & 1 & 1 & 0 & 0 & 1 & 1 & 1 & 1 & 0 \\
g & 1 & 0 & 1 & 0 & 1 & 1 & 0 & 0 & 0 & 1 & 0 & 1 & 0 & 1 & 1 \\
h & 1 & 1 & 0 & 1 & 0 & 1 & 0 & 0 & 0 & 1 & 1 & 0 & 1 & 0 & 1 \\
k & 0 & 1 & 1 & 1 & 1 & 0 & 0 & 0 & 0 & 0 & 1 & 1 & 1 & 1 & 0 \\
\bar{a} & 0 & 0 & 0 & 0 & 0 & 0 & 1 & 1 & 0 & 0 & 1 & 1 & 1 & 1 & 2 \\
\bar{b} & 0 & 0 & 0 & 2 & 0 & 1 & 0 & 1 & 1 & 1 & 0 & 1 & 0 & 1 & 0 \\
\bar{c} & 0 & 0 & 0 & 0 & 2 & 1 & 1 & 0 & 1 & 1 & 1 & 0 & 1 & 0 & 0 \\
\bar{d} & 0 & 2 & 0 & 0 & 0 & 1 & 0 & 1 & 1 & 1 & 0 & 1 & 0 & 1 & 0 \\
\bar{e} & 0 & 0 & 2 & 0 & 0 & 1 & 1 & 0 & 1 & 1 & 1 & 0 & 1 & 0 & 0 \\
\bar{f} & 0 & 1 & 1 & 1 & 1 & 0 & 1 & 1 & 0 & 2 & 0 & 0 & 0 & 0 & 0
\end{array} = R(123,455) = \tilde{C} \times R \times C$$

This representation is analogous to the topological description which has already been given for the same system[5], the non-zero elements being the turning-points (or transition states) to go from one isomer to another.

The 123,455 system may analogously be simplified further on[8] for instance by setting 4=3; one gets

$$\begin{array}{c|cccccccc|}
 & a & b & c & f & g & k & \bar{b} & \bar{c} \\
\hline
a & 0 & 1 & 1 & 2 & 2 & 0 & 1 & 1 \\
b & 1 & 0 & 3 & 1 & 1 & 1 & 1 & 0 \\
c & 1 & 3 & 0 & 1 & 1 & 1 & 0 & 1 \\
f & 2 & 1 & 1 & 0 & 2 & 0 & 1 & 1 \\
g & 2 & 1 & 1 & 2 & 0 & 0 & 1 & 1 \\
k & 0 & 1 & 1 & 0 & 0 & 0 & 1 & 1 \\
\bar{b} & 1 & 1 & 0 & 1 & 1 & 1 & 0 & 3 \\
\bar{c} & 1 & 0 & 1 & 1 & 1 & 1 & 3 & 0
\end{array} = R(123,355)$$

which is the topological matrix of a somewhat modified seven-dimensional tetrahedron[7]: five edges are missing ($r_{ak} = r_{b\bar{c}} = r_{c\bar{b}} = r_{fk} = r_{gk} = 0$ instead of 1), three edges are twofold degenerate ($r_{af} = r_{ag} = r_{fg} = 2$ instead of 1), two edges are threefold degenerate ($r_{bc} = r_{\bar{b}\bar{c}} = 3$ instead of 1). One gets thus a matrix representation which is similar to the topological description given before[6].

Example 2: $12\overline{3,3}55$. The introduction of a chemical bond between ligands 3 and 3 for instance reduces the number of isomers, which can be seen on the three matrices P. One gets a=k=0. If one wants to obtain from $R(123,355)$, the matrix $R(12\overline{3,3}55)$ describing this chelate case, one has

$$\begin{bmatrix} 0&1&0&0&0&0&0&0 \\ 0&0&1&0&0&0&0&0 \\ 0&0&0&1&0&0&0&0 \\ 0&0&0&0&1&0&0&0 \\ 0&0&0&0&0&0&1&0 \\ 0&0&0&0&0&0&0&1 \end{bmatrix} \times R(123,355) \times \begin{bmatrix} 0&0&0&0&0&0 \\ 1&0&0&0&0&0 \\ 0&1&0&0&0&0 \\ 0&0&1&0&0&0 \\ 0&0&0&1&0&0 \\ 0&0&0&0&0&0 \\ 0&0&0&0&1&0 \\ 0&0&0&0&0&1 \end{bmatrix} = \begin{array}{c|cccccc|} & \mathrm{b} & \mathrm{c} & \mathrm{f} & \mathrm{g} & \bar{\mathrm{b}} & \bar{\mathrm{c}} \\ \mathrm{b} & 0&3&1&1&1&0 \\ \mathrm{c} & 3&0&1&1&0&1 \\ \mathrm{f} & 1&1&0&2&1&1 \\ \mathrm{g} & 1&1&2&0&1&1 \\ \bar{\mathrm{b}} & 1&0&1&1&0&3 \\ \bar{\mathrm{c}} & 0&1&1&1&3&0 \end{array}$$

$R(12\overline{3,3}55)$ also remembers the topological matrix of a five-dimensional tetrahedron: two edges are missing ($\mathrm{b} \neq \bar{\mathrm{c}}$ and $\mathrm{c} \neq \bar{\mathrm{b}}$), two edges are threefold degenerate (r_{cb} and $r_{\bar{\mathrm{c}}\bar{\mathrm{b}}}$), one is twofold degenerate (r_{fg}). One gets thus a description which is, once more, analogous to the topological representation given before[6].

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