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ON THE ORDERING OF KEKULÉ STRUCTURES,

Spectral moments as measures of conjugation in cyclobutadiene derivatives

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Abstract

The Kekulé VB structures of a group of benzenoid hydrocarbons containing four-membered rings are transformed into the subspace of their double bonds to produce the corresponding submolecule graphs[7]. Spectral moments are computed for these graphs up to eighth power. Comparability conditions of Miurhead are used to order the partial sums resulting from spectral moment codes. The resulting orders reproduce orders of Kekulé structures based on their Kekulé indices[2], and conjugated circuits[6] within certain defined intervals of submolecules. Mathematical properties of spectral moments of this class of compounds are introduced and a relation to Clar sextet theory[14, 15] is given.

INTRODUCTION

At the beginning of this century, Fries[1], on purely empirical grounds, formulated his famous qualitative rule which states that the most stable Kekulé type structures are those with maximum number of benzenoid rings. Many years later, a group of graph-theorists[2] assigned indices to the individual Kekulé structures, called Kekulé indices, K(L), by projecting occupied MO's on a space defined by functions which characterize individual C=C bonds, selected to

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correspond to formal valence structure to be weighed relative to other Kekulé structures in the set. Let N be the number of C=C double bonds in a particular VB Kekulé structure, then its K(L) is approximately given by eqn. (1), viz.,

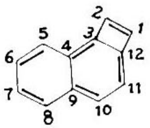
$$K(L) = \frac{1}{2N} \sum_{(i,j)\in L} (q_i + q_j + 2p_{ij})^{\frac{1}{2}} \qquad (1)$$

where q's are charge densities and p's are bond orders. The summation includes only atoms i and j which are joined by double bonds in the given Kekulé structure, L. This index has so far proved consonant with Fries rule[1] and with experimental facts[2,3]; furthermore it illustrates an overlap between MO and VB theories[4]. Since graph-theoretical, G.T., invariants are essentially VB characters, one is tempted to associate a G.T. invariant to set of Kekulé structures and see if it reproduces the same <u>order</u> as predicted from their K(L) indices. There are three interesting aspects of this problem, viz. i) Its MO-VB implications, ii) It deals with the subject of ordering of graphs[5] and iii) It requires certain novel codings of the individual VB structures. The revival of interest in VB methods[4] using powerful mathematical apparatus of graph theory, together with the importance of cyclobutadiene derivatives from both theoretical and experimental interests[3] justify this work.

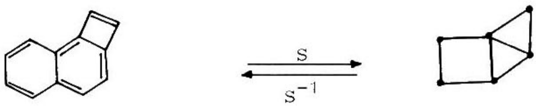
GRAPH-THEORETICAL METHOD

1. Codification of Kekulé structures:

Ordering a set of graphs requires carrying out certain comparability tests[5] (see later) on certain sequences of numbers which might be associated with each graph. Since Kekulé structures are no more than double bond permutations[4], one way to code them is to list a series of numbers representing the positions of double bonds in the relevant structure[3], E.g. the following VB structure would have the code (1-2, 3-4, 5-6, 7-8, 9-10, 11-12). Such a code,



however will depend on the way of labeling of the Kekulé structure and so cannot be regarded as an invariant of the permutations of double bonds. Another approach to coding Kekulé structures makes use of the theory of conjugated circuits of Randić[6]. To do this, however, one must deal with Kekulé structures having only one type of conjugated circuits, e.g. R_m, standing for circuit sizes of 4m+2 pi electrons or Q_m for 4m-circuits. In either case one lists the numbers of circuits of different sizes in an ascending order. Since our cyclobutadienes possess two types of circuits, viz. R_m and Q_m this method cannot be adopted. Therefore one must look for a way of converting double-bond permutations into connected graphs; in which case the topology of the resulting graphs, representing the set of VB structures of a hydrocarbon, will be invariant of the particular permutation. A simple way of doing so is to replace double bonds by vertices, annihilate single bonds, and connect any two vertices that correspond to two double bonds separated by one single bond in the original L(≡K). Thus, e.g.



K S(K)

It seems that Joela[7] was the first to generate these graphs and called them submolecule graphs, S(K), since they result when a Kekulé structure is transformed into the subspace of its double bonds. A relation has been recently established, by one of the authors[8], between connectivities[9] of such

S(K)'s and K(L) values of the corresponding VB structures. We now postulate that the MO characters of the individual Kekulé structures are reproduced by the G.T. characters of S(K) graphs associated with them. Naturally the S(K)'s are quite convenient representatives of the mathematical states which the VB structures describe, and thus any code which might be associated with an S(K) will also represent the individual L which generated that S(K). We choose in this work self-returning walks to code our S(K)'s (and whence their corresponding Kekulé VB structures). There are at least two reasons for our choice, viz., a) Random walks are easy quantities to compute, since they involve only matrix multiplications, b) These quantities have been extensively used in physics and biology[10], but so far only once (as far as we are concerned) in chemistry[11].

2. Computation of self-returning walks of S(K)'s:

First we define[11] a random walk in a graph as a sequence of edges which can be continuously traversed, starting from any vertex and ending on any vertex and allowing repeated use of the same edge or edges. When the random walk starts and ends at the same vertex it is called a self-returning walk. Such walks are easy to enumerate by considering different powers of the adjacency matrix, $\underline{A}$ of the S(K) graph. Thus the elements of the matrix $\underline{A}^k$ are interpreted as walks of length k. In this work we computed $\underline{A}$, $\underline{A}^2$, ... $\underline{A}^m$, (m = 8). The trace of $\underline{A}^k$, tr$\underline{A}^k$, (i.e. sum of its diagonal elements, k being an integer) are called spectral moments of the graph and will be designated here as s_1, s_2, ... s_m. (Where s_i = tr $\underline{A}^i$, i = 1, 2, ...). Thus every S(K) will be coded by a sequence of integers: (s_1, s_2, ... s_8) which we will assume to reflect the mathematical state represented by the corresponding L.

3. Comparability tests of S(K) graphs:

Let S(K) and S(K') be two submolecule graphs to be compared and ordered; and let their spectral moments be defined by the sequences:

$(s_1, s_2, \ldots, s_8)$ and $(s_1', s_2', \ldots, s_8')$ respectively.

According to Muirhead[5] S(K) is comparable with and precedes S(K') only if:

$$\begin{aligned} s_1 &\geq s_1' \\ s_1 + s_2 &\geq s_1' + s_2' \\ &\vdots \\ \sum_{i=1}^{m} s_i &= \sum_{i=1}^{m} s_i' \end{aligned}$$

The last equality, however, is optional[12].

RESULTS

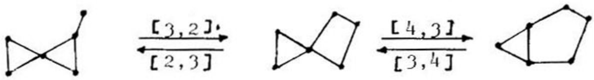
FIG 1 shows S(K)'s studied in this work.

TABLE 1 contains the first eight spectral moments of the S(K)'s, molecular resonance energies of the individual VB structures based on conjugated circuits, and their K(L) indices. The table also lists number of edges, e, in each S(K).

To discuss some of the mathematical properties of this class of benzenoid hydrocarbons containing four membered rings we introduce the concept of a degenerate transformation within a set of S(K) graphs:

Definition

An edge in an S(K) is characterised as an (i,j)-edge where i and j are degrees[13] (i.e. valencies) of its two vertices. A degenerate transformation, $\hat{\Upsilon}$ is defined here as rearrangement of an (i,j)-edge from vertex j to an adjacent vertex k to produce another S(K) belonging to the same hydrocarbon. (a vertex j is one the degree of which is j, and so on). A $\hat{\Upsilon}$ which changes an (i,j)-edge into an (i,k) edge will be denoted as [j,k] transformation. Examples of degenerate transformations in naphtho[a] cyclobutadiene are the following;

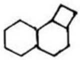


1b 1c 1d

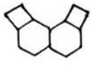
where the alphanumerics of the S(K)'s correspond to those of FIG 1. One observes that 1a is not related to any of the rest of S(K)'s of this hydrocarbon by a $\hat{\Upsilon}$.

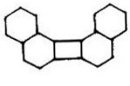
MATHEMATICAL PROPERTIES OF SPECTRAL MOMENTS OF BENZENOID HYDROCARBONS CONTAINING FOUR-MEMBERED RINGS:

First we observe that the set of S(K)'s belonging to a particular topology i.e. corresponding to the VB states of a hydrocarbon may be, arbitrarily, subdivided into subsets, such that the members of each subset are interconvertible to one another by an [i,j] $\hat{\Upsilon}$. We illustrate this observation using the graphs studied and shown in FIG 1.

 → {1a} , {1b, 1c, 1d}

 → {2a, 2b} ; {2c, 2d, 2e}

 → {3a, 3b}; {3c, 3d, 3e}

 → {4a, 4b, 4c}; {4d, 4e, 4f};
{4g, 4h}; {4i}

The members in braces are interconvertible to one another by degenerate transformations. Within our limited sample of studied S(K)'s one observes that members which are degenerate transformates of one another have identical number of edges and their corresponding VB Kekulé structures have identical numbers of conjugated circuits (C.f. Table 1). The above observations are useful in studying the mathematical properties of the spectral moments of this class of hydrocarbons. First we list and prove some properties of the self returning walks that are general to all graphs (properties 1-4) and then we cite three properties specific to this class of Kekulé structures (properties 5-7).

1° <u>s_i ($i \neq 1$) is always an even number</u>. This property might be proved by considering the sum of the diagonal elements of matrices raised to various powers. Let $a^k_{ii} = (A^k)_{ii}$ be a diagonal matrix element in $\underline{A}^k$ (i.e. a^k_{ii} is the number of self returning walks from vertex i to itself possessing length k). As an illustration we consider k = 2. The following expressions might be written for the diagonal elements in $\underline{A}^2$:

$$(A^2)_{11} = (a_{11})^2 + (a_{12})^2 + \ldots + (a_{1n})^2$$
$$(A^2)_{22} = (a_{21})^2 + (a_{22})^2 + \ldots + (a_{2n})^2$$
$$\vdots$$
$$(A^2)_{nn} = (a_{n1})^2 + (a_{n2})^2 + \ldots + (a_{nn})^2$$

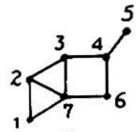
where n is the dimension of $\underline{A}$. Furthermore since $\underline{A}$ is a symmetric matrix in which, by definition, $a_{ii} = 0$; $1 \leqslant i \leqslant n$, the above equations when summed take the following form:

$$\mathrm{tr}\, \underline{A}^2 = s_2 = \sum_{i=1}^{n} (A^2)_{ii} = 2(a_{12})^2 + 2(a_{13})^2 + \ldots + 2(a_{1n})^2 + 2(a_{23})^2 + \ldots + 2(a_{2n})^2 + \ldots + 2(a_{n-1,n})^2 .$$

Thus s_2 is the sum of pairs of terms, whence it should be even. This result is general and might be proved for higher values of k in a similar way.

2° a_{ii}^k is always even for odd values of k

The reverse is not true, however, i.e. a_{ii}^k is not necessarily odd for even values of k. To illustrate this property we list a_{ii}^k values for the first eight powers of $\underline{A}$ for graph 2a:



2a

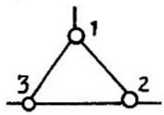
i	a_{ii}	a_{ii}^2	a_{ii}^3	a_{ii}^4	a_{ii}^5	a_{ii}^6	a_{ii}^7	a_{ii}^8
1	0	2	2	11	20	79	178	616
2	0	3	4	17	40	127	344	1025
3	0	3	2	20	26	145	260	1102
4	0	3	0	14	4	84	68	571
5	0	1	0	3	0	14	4	84
6	0	2	0	11	6	72	82	513
7	0	4	4	26	44	193	408	1503
$\sum$:	0	18	12	102	140	714	1344	5414

where the last row is the sequence of s_i's, $1 \leq i \leq 8$ i.e. the code of this submolecule.

To prove this conjecture we consider each of two possible cases separately. In the first case S(K) contains no odd-membered cycles, and consequently $a_{ii}^k = 0$ for k odd. In the second case, S(K) contains some number of odd-membered cycles. A self returning walk of the odd length k starting from vertex i and ending at the same vertex implies it goes over some (odd) number of odd-membered cycles. As any cycle could be passed in two oppositely oriented ways the above self returning walks appear always in pairs. This proves the conjecture for both cases.

3° Let e = number of edges in the graph, then $\underline{s_2 = 2e}$. This fact results because every edge has two vertices, and every vertex contributes one self returning walk of length two per edge , thus every edge contributes two such walks.

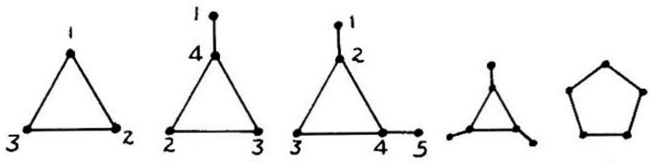
4° Let nC_3 be the number of 3-membered cycles in the graph, then $\underline{s_3 = 6nC_3}$. E.g. nC_3 (2b) = 2, s_3 (2b) = (6)(2) = 12.
This property is proved as follows: Consider a C_3 in an arbitrary graph



First one observes that s_3 results <u>only</u> if the graph contains one or more C_3's. Each C_3 leads to <u>six</u> self returning walks of length three each, viz. (123), (132); (213), (231); (312) and (321). This demonstrates the property.

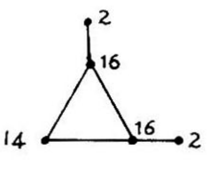
5° <u>The values of s_5 are always integral multiples of ten</u>.

<u>Proof</u>: First we observe that s_5 results from one or more of the following graphs:

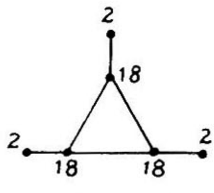


C_3 g_1 g_2 g_3 C_5

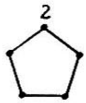
It can easily be shown that each of the above subgraphs leads to a number of s_5's which is a multiple of 10. Thus each of the vertices in C_3 contributes 10 self returning walks of length 5 each. These walks for v_1, e.g., are: (123131), (132131), (131231), (131321), (123121), (132121), (121321), (121231), (123231), and (132321). Thus each C_3 denotes 30 s_5's. Similarly v_4 in g_1 contributes 14 self returning walks to s_5. Ten of these walks are the "endocyclic" ones of the C_3 in addition to 4 other walks, viz. (414234), (414324), (423414), and (432414). Each of v_2 and v_3 donates 12 walks of length five. These are 10 endocyclic ones and two others involving v_1, e.g. these two walks for v_3 are (341423) and (324143). The exocyclic vertex, v_1 gives two walks of length 5, viz. (142341) and (143241). Thus g_1 contains a total of 14+12+12+2 = 40 walks of length 5 each = s_5. We can easily compute self returning walks of length 5 for each of g_2, g_3 and C_5. Such walks are assigned to relevant vertices as shown below



$s_5 = 2(2) + 14 + 2(16) = 50$



$s_5 = 3(2) + 3(18) = 60$



$$s_5 = 2(5) = 10$$

These walks might be computed by considering the diagonal elements of the corresponding $\underline{A}^5$ matrices or inferred directly from the relevant graphs. E.g. v_2 in g_2 is involved in 16 self-returning walks of length 5 each. Ten of these walks are the endocyclic ones, while the additional six walks are: (212342), (212432), (245432), (234212), (243212) and (234542). Thus, since each of the subgraphs involved in s_5 generate numbers of self returning walks of length five which are multiples of ten, the propery is proved.

6° Let $\{S(K_i), S(K_j), S(K_k), \ldots\}$ be a subset of degenerate transformates i.e. each S(K) is transformable to any other member of the subset by one or more $\hat{\mathcal{T}}$ operations.
Let $|\Delta s_i|$ = absolute value of difference between any two s_i values of two S(K)'s of the same subset. The following observatioh is conjectured:

$$|\Delta s_i| = in; \; (n = 0, 1, 2, \ldots ; \; i = 1, 2, \ldots , 7)$$

We might illustrate the above relation by examining the subsets of 1, 2, 7, 8-dibenzobiphenylene, (4a-4i) of FIG 1 and TABLE 1. Spectral moment codes are given below:

	s_1	s_2	s_3	s_4	s_5	s_6	s_7
4a:	0	26	24	122	220	746	1708
			↓ 3x2 ↑	↓ 4 ↑	↓ 5x10 ↑	↓ 6 ↑	↓ 7x54 ↑
4b:	0	26	18	126	170	752	1330
			↓ 3x2 ↑	↓ 4 ↑	↓ 5x10 ↑	↓ 6 ↑	↓ 7x54 ↑
4c:	0	26	12	130	120	758	952

	s_1	s_2	s_3	s_4	s_5	s_6	s_7
4d:	0	26	12	138	100	890	784
				4x3	5x2	6x24	7x14
4e:	0	26	12	126	110	746	882
				4x3	5x2	6x22	7x10
4f:	0	26	12	114	120	614	952
				(4x11.5)		(6x97.333)	
4g:	0	28	24	160	280	1198	2786
				4	5x12	6x2	7x80
4h:	0	28	18	164	220	1186	2226
4i:	0	28	12	164	180	1162	1988

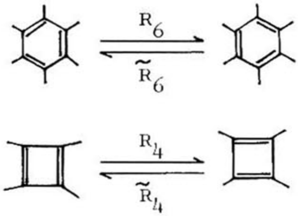
The above type of relation is symptomatic of other relations between code integers of other S(K)'s studied in this work.

7° Spectral moment of S(K) and Kekulé index of the corresponding Kekulé VB structure:
Investigation of Table 1 makes it clear that the order of spectral moment codes reproduces the order of K(L) values within subsets of degenerate transformates.
FIG 2 is a hierarchical diagram of the S(K)'s defining compound 4, when conditions of Muirhead are applied to their spectral moment codes. The resulting order also reproduces the order of molecular resonance energy per Kekulé structure (based on conjugated circuits[6]).

8° Relation to Clar's sextet theory[14]

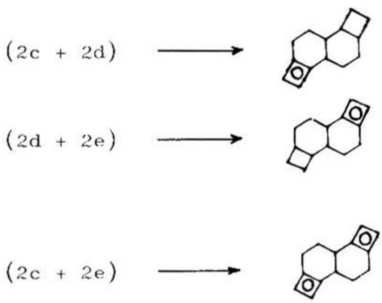
Recently, Hosoya et al.[15] presented a graph-theoretical analysis of Clar's sextet theory. Their analysis was, however, confined only to polyhex graphs. We introduce the following definitions to include benzenoid hydrocarbons containing four-membered rings:

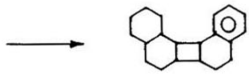
R_n transformation (n = 6, 4):

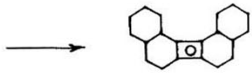


Superposition of two Kekulé structures

In a set of Kekulé structures belonging to a hydrocarbon two structures might differ only in the orientation of just one sextet or one quartet of electrons. According to Clar one can draw a circle representing the sextet or quartet in the hexagon or square (respectively) concerned. If furthermore double bonds are converted into single bonds one obtains various sextet and quartet patterns. E.g.



(4a + 4b) 

(4a + 4d) 

One observes that when two Kekulé structures are related to one another by an R_6 or R_4 (or the inverse operators, $\tilde{R}_6$, $\tilde{R}_4$), their submolecule graphs will be degenerate transformates of one another i.e.

$$\text{if} \quad R_n K_i = K_j \quad ; \quad \tilde{R}_n K_j = K_i \qquad (n = 4;\ 6)$$

$$\text{then} \quad S(K_i) \underset{}{\overset{\hat{\tau}}{\rightleftharpoons}} S(K_j)$$

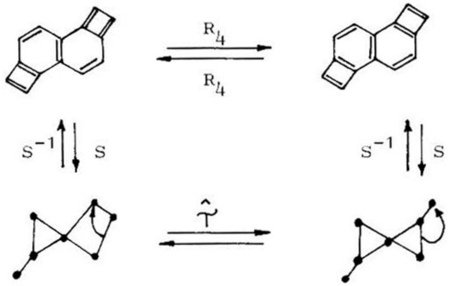
where $\hat{\tau}$ is a degenerate transformation operator defined in terms of the R_n (and $\tilde{R}_n$) operators as follows:

$$(1) \quad S(K_i) \xrightarrow{S^{-1}} K_i$$

$$(2) \quad K_i \xrightarrow{R_n \text{ or } \tilde{R}_n} K_j \quad ; \quad (n = 4,\ 6)$$

$$(3) \quad K_j \xrightarrow{S} S(K_j)$$

where S^{-1} converts an $S(K)$ into its corresponding K. i.e. $\hat{\tau} \equiv (S^{-1};\ R_n\ (\text{or } R_n);\ S)$. As an illustration we consider the following transformations:

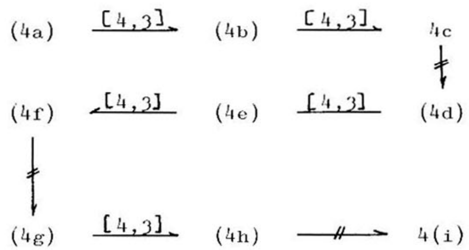


<u>Conjecture</u>

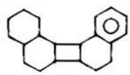
When two Kekulé structures are related by an R_n or $\tilde{R}_n$ (n = 4, 6) they will have adjacent characters (i.e. adjacent values of K(L) and conjugated circuits), in which case, their submolecules will be degenerate transformates of one another and will have adjacent spectral moment codes.

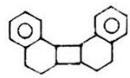
Illustrations:

The following $\hat{\mathcal{T}}$ operations are defined among the submolecules of hydrocarbon 4 (FIG 1),



One observes that the values of K(L) and conjugated circuits of Kekulé structures corresponding to $\{(4a), (4b)$ and $(4c)\}$ are adjacent (C.f. Fig 2). A similar situation occurs with subsets of $\{4f, 4e, 4d\}$ and $\{4g, 4h\}$. The elements of each subset generate adjacent codes. Furthermore, two adjacent VB structures of each subset gives rise to a Clar formula with <u>one</u> circle when superposed, thus:

(4a + 4b) ⟶ 

but (4a + 4c) ⟶ 

So we conclude a hierarchy of characters 4a ⟶ 4b ⟶ 4c (but not 4a ⟶ 4c ⟶ 4b). Thus the number of Clar's circles resulting when two VB states are combined gives an indication of their relative positions. Thus two adjacent states lead to only one circle. Other illustrations might be infered from TABLE 1 and FIG. 1.

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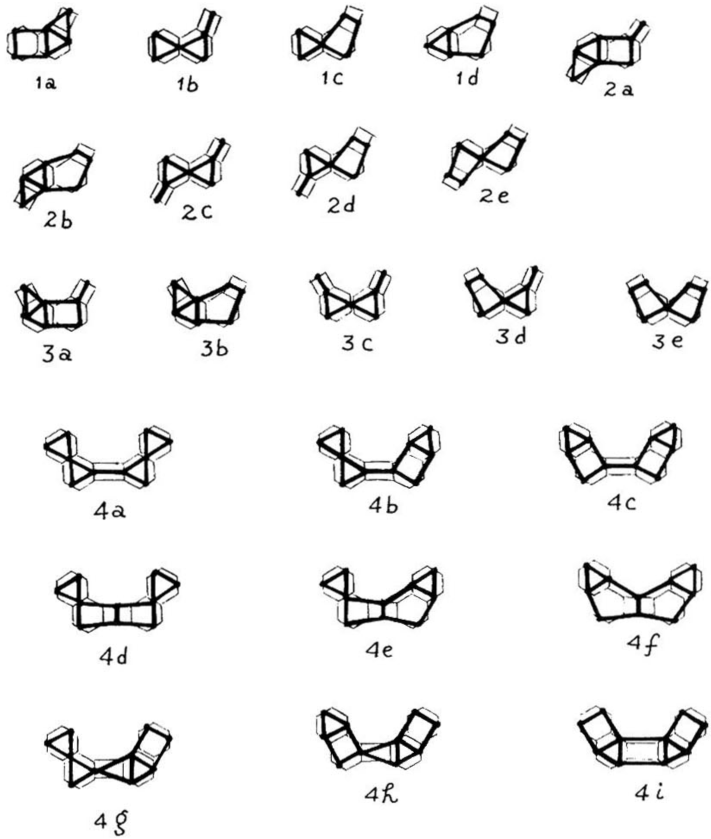

Fig. 1

Submolecule graphs studied in this work. They are heavily drawn inside the molecular graphs of the corresponding hydrocarbons. Relevant graph-theoretical data are found in Table 1.

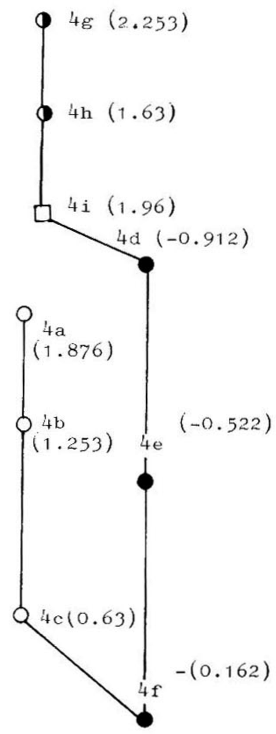

Fig. 2

Hierarchical diagram of S(K)'s of compound 4 when conditions of Muirhead are imposed on partial sums of their spectral moments (up to $\underline{A}^8$). The resulting order reproduces molecular resonance energy based on conjugated circuits (numbers in parentheses, Table 1). Only sets of identical "colors" are to be compared (O: cc = 5, e = 13; ●: cc = 6, e = 13; ◐: cc = 6, e = 14; □: R = 8, e = 14) (cc = number of conjugated circuits in Kekulé structure, e = number of edges in the corresponding S(K)).

TABLE 1

Spectral moments of submolecules and MO-VB characters of Kekulé structures

S(K)	Spectral moments and partial sums	Conjugated circuits* (eV)	K(L)*	e**
1a	0, 16, 12, 92, 140, 646, 1316, 4868 0, 16, 28, 120, 260, 609, 2222, 7090	$R_1+R_2+Q_2+Q_3 = 0.515$	0.926	8
1b	0, 14, 12, 62, 110, 362, 826, 2358 0, 14, 26, 88, 198, 560, 1386, 3744	$2R_1+Q_1 = 0.138$	0.916	7
1c	0, 14, 6, 66, 50, 362, 378, 2082 0, 14, 20, 86, 136, 498, 878, 2958	$R_1+Q_2+Q_3 = 0.269$	0.860	7
1d	0, 14, 6, 54, 60, 260, 434, 1390 0, 14, 20, 74, 134, 394, 828, 2218	$R_1+Q_1+Q_3 = -0.881$	0.845	7
2a	0, 18, 12, 102, 140, 714, 1344, 5414 0, 18, 30, 132, 272, 986, 2330, 7744	$R_1+R_2+Q_1+Q_2+Q_3 = -1.085$	0.8798	9
2b	0, 18, 12, 90, 140, 582, 1274, 4226 0, 18, 30, 120, 260, 842, 2116, 6342	$R_1+R_3+Q_1+Q_2+Q_3 = -1.231$	0.8897	9
2c	0, 16, 12, 72, 120, 424, 952, 2832 0, 16, 28, 100, 220, 644, 1596, 4428	$2R_1+2Q_1 = -1.462$	0.8599	8
2d	0, 16, 6, 76, 60, 424, 490, 2532 0, 16, 22, 98, 158, 582, 1072, 3604	$R_1+2Q_1+Q_2 = -2.781$	0.8697	8

TABLE 1 (Cont.)

S(K)	Spectral moments and partial sums	Conjugated circuits* (eV)	K(L)*	e**
2e	0, 16, 0, 80, 0, 448, 0, 2624 / 0, 16, 16, 96, 96, 544, 544, 3168	$2Q_1+2Q_2$ = -4.1	0.8796	8
3a	0, 18, 12, 102, 140, 720, 1358, 5486 / 0, 18, 30, 132, 272, 992, 2350, 7836	$R_1+R_2+Q_1+Q_2+Q_3$ = -1.085	0.8870	9
3b	0, 18, 12, 90, 140, 582, 1274, 4226 / 0, 18, 30, 120, 260, 842, 2116, 6342	$R_1+R_3+Q_1+Q_2+Q_3$ = -1.231	0.8871	9
3c	0, 16, 12, 72, 120, 424, 952, 2832 / 0, 16, 28, 100, 220, 644, 1596, 4428	$2R_1+2Q_1$ = -1.462	0.8687	8
3d	0, 16, 6, 76, 60, 424, 490, 2532 / 0, 16, 22, 98, 158, 582, 1072, 3604	$R_1+2Q_1+Q_2$ = -2.781	0.8688	8
3e	0, 16, 0, 80, 0, 448, 0, 2624 / 0, 16, 16, 96, 96, 544, 544, 3168	$2Q_1+2Q_2$ = -4.1	0.8689	8
4a	0, 26, 24, 122, 220, 746, 1708, 5066 / 0, 26, 50, 172, 392, 1138, 2846, 7912	$4R_1+Q_1$ = 1.876	0.9071	13
4b	0, 26, 18, 126, 170, 752, 1330, 4862 / 0, 26, 44, 170, 340, 1092, 2422, 7284	$3R_1+R_2+Q_1$ = 1.253	0.8990	13

TABLE 1 (Cont.)

S(K)	Spectral moments and partial sums	Conjugated circuits* (eV)	K(L)*	e**
4c	0, 26, 12, 130, 120, 738, 952, 4690 0, 26, 38, 168, 288, 1046, 1998, 6688	$2R_1+2R_2+Q_1 = 0.63$	0.8909	13
4d	0, 26, 12, 138, 100, 890, 784, 6154 0, 26, 38, 176, 276, 1166, 1950, 8104	$2R_1+Q_1+2Q_2+Q_3 = -0.912$	0.8860	13
4e	0, 26, 12, 126, 110, 746, 882, 4798 0, 26, 38, 164, 274, 1020, 1902, 6700	$2R_1+Q_1+Q_2+Q_3+Q_4=-0.522$	0.8779	13
4f	0, 26, 12, 114, 120, 614, 952, 3674 0, 26, 38, 152, 272, 886, 1838, 5512	$2R_1+Q_1+2Q_3+Q_5 = -0.162$	0.8698	13
4g	0, 28, 24, 160, 280, 1198, 2786, 10104 0, 28, 52, 212, 492, 1690, 4476, 14580	$3R_1+R_2+Q_2+Q_3 = 2.253$	0.9080	14
4h	0, 28, 18, 164, 220, 1186, 2226, 9484 0, 28, 46, 210, 430, 1616, 3842, 13326	$2R_1+2R_2+Q_2+Q_3 = 1.63$	0.8998	14
4i	0, 28, 12, 164, 180, 1162, 1988, 9188 0, 28, 40, 204, 384, 1546, 3534, 12722	$2R_1+2R_2+Q_3+2Q_4+Q_5=1.96$	0.9088	14

* Of the corresponding Kekulé structure

** Number of edges in S(K)