

# Aromaticity and Diatropicity of Polyacenequinododimethides

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**Abstract:** Typical polyacenequinododimethides exist only in a single classical structure. These hydrocarbons are moderately aromatic and diatropic, although they have no aromatic conjugated circuits. This apparent dichotomy was resolved with our graph theory of aromaticity and magnetotropy. Many nonconjugated circuits were found to contribute collectively to aromaticity and diatropicity. For individual molecules, local aromaticity increases with distance from the *exo*-methylene groups. This fact indicates that the conjugated-circuit model is not always applicable to semibenzenoid hydrocarbons such as polyacenequinododimethides.

**Keywords:** aromaticity • magnetic properties • polyacenequinododimethides • polyacenes • ring currents

## Introduction

The conjugated-circuit theory states that conjugated circuits are the main origin of aromaticity in polycyclic aromatic hydrocarbons.<sup>[1,2]</sup> Dewar resonance energies (DREs) of polycyclic benzenoid hydrocarbons can be reproduced well in terms of conjugated circuits. Nonconjugated circuits have been regarded as minor contributors to global aromaticity. In 1975, we tacitly assumed that polyacene-2,3-quinododimethides (2,3-dimethylene-2,3-dihydropolyacenes **7–12**; Figure 1) are olefinic in nature, because only one classical resonance structure can be written for them.<sup>[3]</sup> No conjugated circuits can be chosen from these hydrocarbons. Schleyer and Puhlhofer<sup>[4]</sup> and Hajgató et al.<sup>[5]</sup> used such hydrocarbons with exocyclic methylenes as polyene-like reference structures in homodesmotic reactions.

In 1973, however, Gleicher et al. found that a series of polyacene-2,3-quinododimethides might be appreciably aromatic with positive Hess–Schaad and Dewar resonance energies.<sup>[6]</sup> Although these hydrocarbons can exist in only a

single classical resonance structure, there is a steady increase in resonance energy per  $\pi$  electron on going to higher members. As a result, pentacene-2,3-quinododimethide (2,3-dimethylene-2,3-dihydropentacene, **11**; Figure 1) has about half the aromatic character associated with pentacene itself.<sup>[6]</sup> Thus, it has been strongly suggested that even polycyclic semibenzenoid hydrocarbons that consist of essential single and double bonds might be aromatic if they are sufficiently large in molecular size.

Recently, Wannere et al. predicted that fulminene-2,10-quinododimethide (2,10-methylene-2,10-dihydrofulminene), which also exists only in a single classical resonance structure, must have <sup>1</sup>H NMR chemical-shift values in the aromatic range.<sup>[7]</sup> They nevertheless viewed this hydrocarbon as an essentially nonaromatic species. Faglioni et al. pointed out soon afterward that this hydrocarbon should rather be regarded as an aromatic species, because it sustains a net diatropic  $\pi$  current.<sup>[8]</sup> In this paper, we interpret the peculiar aromatic and diatropic character of polyacenequinododimethides, a prototypical series of quinododimethides, by using our graph theory of aromaticity and magnetotropy<sup>[9–23]</sup> and compare it with that of polyacenes. Magnetotropy refers collectively to diatropicity and paratropicity.

## Theory

Our graph theory of aromaticity and magnetotropy is constructed within the simple Hückel framework. We use the topological resonance energy (TRE) as an indicator of global aromaticity.<sup>[9–11]</sup> Percentage TRE (%TRE) is defined as 100 times TRE divided by the  $\pi$ -binding energy of the

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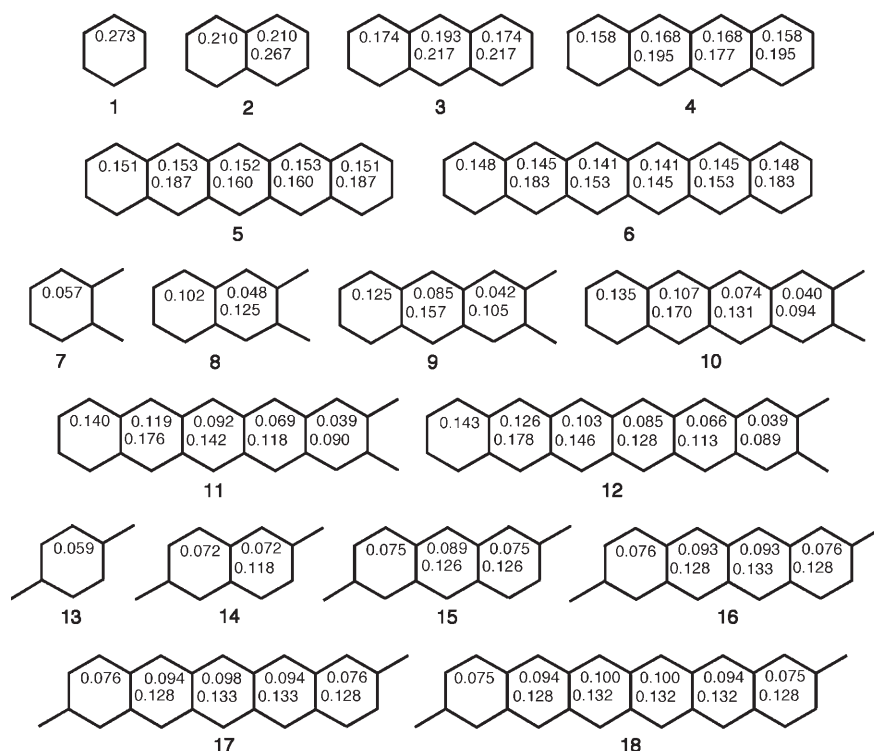


Figure 1. Bond resonance energies (BREs) in units of  $|\beta|$  for all  $\pi$  bonds in polyacenes **1–6** and polyacenequinodimethides **7–18**.

polyene reference.<sup>[11,16,18]</sup> This quantity is used as a measure of aromaticity, normalized with respect to the molecular size. Bond resonance energy (BRE) represents the contribution of a given  $\pi$  bond to the global aromaticity.<sup>[16–19]</sup> Circuits stand for all possible cyclic paths that can be chosen from a cyclic  $\pi$  system.<sup>[24]</sup> For example, there are five-, seven-, and 10-site circuits in azulene. For simplicity, all six-membered rings in all molecules studied are assumed to be regular hexagons in shape.

Circuit resonance energy (CRE) represents an energy gain or loss due to cyclic conjugation along a given circuit.<sup>[20–23]</sup> The CRE for the  $i$ th circuit in a hydrocarbon  $\pi$  system  $G$  is defined as  $A_i$  in the form [Eq. (1)]:<sup>[11,20–23]</sup>

$$A_i = 4 \sum_j^{\text{occ}} \frac{P_{G-r_i}(X_j)}{P_G(X_j)} \quad (1)$$

in which  $r_i$  is a set of carbon atoms that constitute the  $i$ th circuit  $c_i$ ,  $G-r_i$  is the subsystem of  $G$  obtained by deleting

#### Abstract in Japanese:

共鳴構造式が1つしか書けないポリアセンのキノドジメチド類でも、かなり大きな芳香族性を示し、かなり大きな反磁性環電流を誘起する。この現象を相原のグラフ理論を用いて解析したところ、これらの分子では非共役サーキットが芳香族性や反磁性に大きな寄与をすることがわかった。

from  $G$  all carbon atoms that constitute the  $i$ th circuit,  $P_G(X)$  and  $P_{G-r_i}(X)$  are the characteristic polynomials for  $G$  and  $G-r_i$ , respectively, and  $X_j$  is the  $j$ th-largest root of the equation  $P_G(X)=0$ ;  $j$  runs over all occupied  $\pi$  molecular orbitals. If some occupied molecular orbitals have the same energy, this formula must be replaced by others.<sup>[12–14]</sup> The CRE for a six-site circuit correlates well with the so-called six-center index devised by Bultinck.<sup>[25,26]</sup>

The sum of  $A_i$  values or CREs over all circuits represents the aromatic stabilization energy (ASE) for an entire  $\pi$  system. This sum is termed the magnetic resonance energy (MRE), which means a TRE-like ASE derived from the magnetic response of a cyclic  $\pi$  system [Eq. (2)]:<sup>[20–23]</sup>

$$\text{MRE}/|\beta| = \sum_i^G A_i \quad (2)$$

in which  $|\beta|$  is the absolute value of a standard resonance integral between two adjacent carbon  $sp_2$  atomic orbitals.

An excellent correlation is indeed observed between MRE and TRE for polycyclic benzenoid hydrocarbons and heterocycles.<sup>[20–23]</sup> This correlation in turn justifies the interpretation of  $A_i$  values as CREs. Notably, no hypothetical polyene reference is needed to evaluate the MRE.

In fact,  $A_i$  appears in the graph-theoretical formula for ring-current magnetic susceptibility [Eq. (3)]:<sup>[11–13]</sup>

$$\chi_G = 4.5 \chi_0 \sum_i^G A_i \left( \frac{S_i}{S_0} \right)^2 \quad (3)$$

in which  $\chi_0$  is the ring-current susceptibility of benzene and  $S_i$  and  $S_0$  are the areas of  $r_i$  and the benzene ring, respectively. Positive and negative  $A_i$  values represent diamagnetic and paramagnetic contributions, respectively. From this formula, we derive the contribution of the  $i$ th circuit to  $\chi_G$  in the form [Eq. (4)]:<sup>[11–13]</sup>

$$\chi_i = 4.5 \chi_0 A_i \left( \frac{S_i}{S_0} \right)^2 \quad (4)$$

Every circuit is then supposed to sustain a  $\pi$ -electron current in the external magnetic field, the intensity of which is given by [Eq. (5)]:<sup>[13–15]</sup>

$$I_i = 4.5 I_0 A_i \frac{S_i}{S_0} \quad (5)$$

in which  $I_0$  is the intensity of the  $\pi$ -electron current induced in the benzene molecule. Positive and negative  $A_i$  values now represent diatropicity and paratropicity, respectively. Thus, at the level of individual circuits, aromaticity and antiaromaticity correspond without exception to diatropicity and paratropicity, respectively. A total  $\pi$ -current-density map for G is obtained by superposing all the circuit currents. Hückel–London theory<sup>[27,28]</sup> is capable of reproducing the patterns of ab initio  $\pi$ -current density in many polycyclic conjugated hydrocarbons fairly well,<sup>[29,30]</sup> although it is unable to predict the diamagnetic  $\pi$  vortices in the region of the formal C=C bond.<sup>[8]</sup> Our theory of magnetotropy is exactly a variant of the Hückel–London theory.

As has been seen, the total  $\pi$ -current density is dependent on the molecular geometry. To escape the geometric dependence of  $\pi$ -electron currents, hypothetical geometry-independent circuit currents were evaluated by equating all the circuit areas in [Eq. (5)] with that of the benzene ring (i.e.,  $S_i = S_0$  for all circuits).<sup>[20,21]</sup> As will be seen later, the intensity of a geometry-independent  $\pi$  current that is supposed to run through a given peripheral bond, which is obtained by summing the geometry-independent circuit currents that are supposed to run through the bond, is nearly proportional to the corresponding BRE. Thus, the essence of our theory is that the TRE, BRE,  $\pi$ -current density, and ring-current magnetic susceptibility can all be interpreted consistently in terms of CREs.

## Results and Discussion

Six polyacenes **1–6**, six polyacene-2,3-quinododimethides (2,3-dimethylene-2,3-dihydropolyacenes **7–12**), and six polyacene-2, $x$ -quinododimethides (2, $x$ -dimethylene-2, $x$ -dihydropolyacenes **13–18**) were employed for the present study. TREs and MREs for these hydrocarbons are listed in Table 1. As in the case of other polycyclic aromatic hydrocarbons,<sup>[20–22]</sup> the MRE is somewhat smaller than the TRE. For polyacenes, only one sextet ring can be chosen from the  $\pi$  system, so that the aromaticity is gradually diluted on going to higher members.<sup>[31,32]</sup> Therefore, the %TRE decreases on going to higher members with more benzene rings.<sup>[21]</sup> Two series of quinododimethides were tacitly expected to be nonaromatic, as neither sextet rings nor conjugated circuits can be chosen from the  $\pi$  system.<sup>[3–5]</sup> However, all these hydrocarbons proved to be more or less aromatic with smaller positive TREs and MREs. In line with this, some of the quinododimethides have moderately large posi-

Table 1. TREs and MREs for polyacenes and polyacenequinododimethides.

Species	TRE [ $ \beta $ ] <sup>[a]</sup>	MRE [ $ \beta $ ] <sup>[a]</sup>
Polyacenes		
Benzene ( <b>1</b> )	0.2726 (3.528)	0.2222
Naphthalene ( <b>2</b> )	0.3888 (2.924)	0.2894
Anthracene ( <b>3</b> )	0.4746 (2.519)	0.3405
Naphthacene ( <b>4</b> )	0.5531 (2.269)	0.3928
Pentacene ( <b>5</b> )	0.6298 (2.105)	0.4476
Hexacene ( <b>6</b> )	0.7063 (1.992)	0.5041
Polyacene-2,3-quinododimethides:		
Benzene-2,3-quinododimethide ( <b>7</b> )	0.0593 (0.599)	0.0575
Naphthalene-2,3-quinododimethide ( <b>8</b> )	0.1405 (0.913)	0.1297
Anthracene-2,3-quinododimethide ( <b>9</b> )	0.2253 (1.078)	0.1990
Naphthacene-2,3-quinododimethide ( <b>10</b> )	0.3090 (1.169)	0.2639
Pentacene-2,3-quinododimethide ( <b>11</b> )	0.3911 (1.224)	0.3257
Hexacene-2,3-quinododimethide ( <b>12</b> )	0.4717 (1.258)	0.3860
Polyacene-2, $x$ -quinododimethides:		
Benzene-2,4-quinododimethide ( <b>13</b> ; $x=4$ )	0.0612 (0.621)	0.0593
Naphthalene-2,6-quinododimethide ( <b>14</b> ; $x=6$ )	0.1345 (0.877)	0.1247
Anthracene-2,6-quinododimethide ( <b>15</b> ; $x=6$ )	0.2122 (1.018)	0.1895
Naphthacene-2,8-quinododimethide ( <b>16</b> ; $x=8$ )	0.2912 (1.105)	0.2526
Pentacene-2,9-quinododimethide ( <b>17</b> ; $x=9$ )	0.3704 (1.162)	0.3142
Hexacene-2,10-quinododimethide ( <b>18</b> ; $x=10$ )	0.4495 (1.202)	0.3749

[a] The values in parentheses are the %TREs.

tive DREs.<sup>[6]</sup> The %TRE increases upon elongation of the polyacene moiety.

The ring-current diamagnetic susceptibilities calculated for **1–18** are listed in Table 2. All these species are highly or moderately diamagnetic. Each quinododimethide molecule is less diamagnetic than the corresponding polyacene, which suggests that *exo*-methylenes suppress not only cyclic conju-

Table 2. Ring-current diamagnetic susceptibilities for polyacenes and polyacenequinododimethides.

No. of benzene rings	Ring-current diamagnetic susceptibility [ $\chi_0$ ]		
	Polyacene	Polyacene-2,3-quinododimethide	Polyacene-2, $x$ -quinododimethide
1	1.000 ( <b>1</b> )	0.259 ( <b>7</b> )	0.267 ( <b>13</b> ; $x=4$ )
2	2.185 ( <b>2</b> )	0.761 ( <b>8</b> )	0.754 ( <b>14</b> ; $x=6$ )
3	3.448 ( <b>3</b> )	1.441 ( <b>9</b> )	1.406 ( <b>15</b> ; $x=6$ )
4	4.747 ( <b>4</b> )	2.240 ( <b>10</b> )	2.179 ( <b>16</b> ; $x=8$ )
5	6.062 ( <b>5</b> )	3.123 ( <b>11</b> )	3.039 ( <b>17</b> ; $x=9$ )
6	7.385 ( <b>6</b> )	4.067 ( <b>12</b> )	3.965 ( <b>18</b> ; $x=10$ )

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gation but also circulation of  $\pi$  electrons. Ring-current diamagnetic susceptibility and the associated diamagnetic susceptibility exaltation have been used as magnetic indicators of aromaticity.<sup>[33,34]</sup> However, on going to higher members in each series, the susceptibility increases much faster than the TRE or MRE; the susceptibility is never proportional to the TRE or the MRE. It follows that ring-current diamagnetic susceptibility is not a good measure of aromaticity.

The BREs for **1–18** are graphically summarized in Figure 1. The BRE for a peripheral  $\pi$  bond represents the local aromaticity of a ring to which the  $\pi$  bond belongs.<sup>[19]</sup> For polyacene molecules, all the rings are similar in local aromaticity to each other;<sup>[21]</sup> average local aromaticity decreases on increasing the number of benzene rings. For the 2,3-quinododimethide series, local aromaticity is enhanced with distance from the methylene groups; the farthest ring from the *exo*-methylenes in **12** is comparable in local aromaticity to the benzene rings in **6**. For 2,*x*-quinododimethide molecules, the inner benzene rings are more aromatic than the outer ones. The BREs for the *exo*-methylene C–C bonds vanish. As pointed out by Faglioni et al.,<sup>[8]</sup> the perturbation produced by the *exo*-methylenes decrease with distance from the methylene groups. For the physical meaning of local aromaticity, see references [19], [25], and [26].

We now proceed to interpret the aromatic and diatropic character of **1–18** in terms of CREs. Many circuits can be chosen from the polycyclic  $\pi$  systems in **1–18**. As shown in Figure 2, hexacene (**6**) and hexacene-2,10-quinododimethide

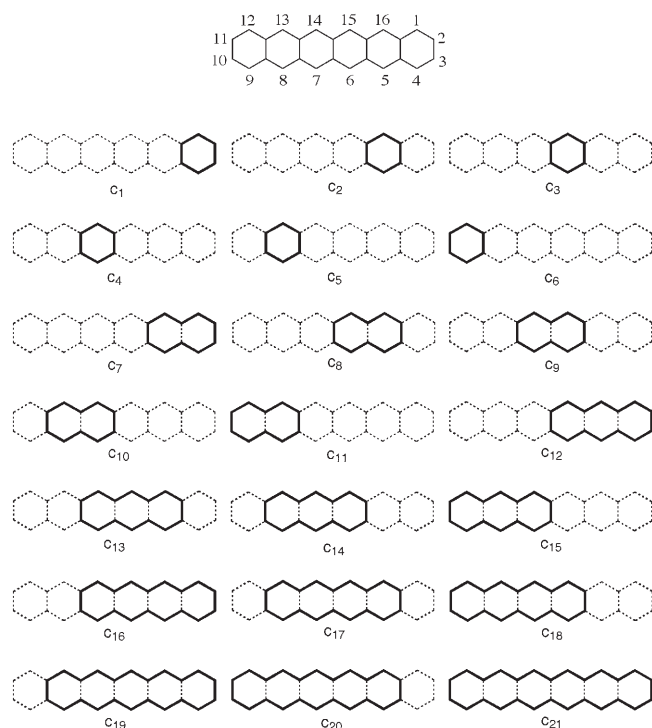


Figure 2. The non-identical circuits in hexacene (**6**), hexacene-2,3-quinododimethide (**12**), and hexacene-2,10-quinododimethide (**18**). Two *exo*-methylenes in **12** and **18** are attached to the 2,3- and 2,10-positions of hexacene, respectively.

(**18**) have 12 non-identical circuits and 21 circuits in all, whereas hexacene-2,3-quinododimethide (**12**) has 21 non-identical circuits and 21 circuits in all. The CREs and intensities of circuit currents for the three benchmark hydrocarbons are listed in Tables 3 and 4, respectively. As for the numbering of circuits in these hydrocarbons, see Figure 2. In brief, all circuits in **1–18** are aromatic and diatropic to varying extents.

We previously discussed the relative magnitudes of CREs for all circuits in polyacenes **1–6**,<sup>[20–22]</sup> all these circuits are

Table 3. CREs for hexacene (**6**) and hexacenequinododimethides **12** and **18**.

Circuit	Area [ $S_0$ ]	CRE [ $ \beta $ ]		
		<b>6</b>	<b>12</b>	<b>18</b>
c <sub>1</sub>	1	0.0855	0.0296	0.0516
c <sub>2</sub>	1	0.0490	0.0395	0.0466
c <sub>3</sub>	1	0.0426	0.0416	0.0444
c <sub>4</sub>	1	0.0426	0.0433	0.0444
c <sub>5</sub>	1	0.0490	0.0506	0.0466
c <sub>6</sub>	1	0.0855	0.0880	0.0516
c <sub>7</sub>	2	0.0237	0.0056	0.0124
c <sub>8</sub>	2	0.0139	0.0088	0.0123
c <sub>9</sub>	2	0.0120	0.0106	0.0120
c <sub>10</sub>	2	0.0139	0.0140	0.0123
c <sub>11</sub>	2	0.0237	0.0250	0.0124
c <sub>12</sub>	3	0.0098	0.0017	0.0045
c <sub>13</sub>	3	0.0068	0.0033	0.0050
c <sub>14</sub>	3	0.0068	0.0053	0.0050
c <sub>15</sub>	3	0.0098	0.0098	0.0045
c <sub>16</sub>	4	0.0057	0.0007	0.0021
c <sub>17</sub>	4	0.0049	0.0019	0.0025
c <sub>18</sub>	4	0.0057	0.0042	0.0021
c <sub>19</sub>	5	0.0045	0.0004	0.0011
c <sub>20</sub>	5	0.0045	0.0016	0.0011
c <sub>21</sub>	6	0.0042	0.0004	0.0005

Table 4. Circuit currents for hexacene (**6**) and hexacenequinododimethides **12** and **18**.

Circuit	Area [ $S_0$ ]	Circuit current [ $I_0$ ]		
		<b>6</b>	<b>12</b>	<b>18</b>
c <sub>1</sub>	1	0.3846	0.1331	0.2324
c <sub>2</sub>	1	0.2204	0.1780	0.2095
c <sub>3</sub>	1	0.1916	0.1874	0.1999
c <sub>4</sub>	1	0.1916	0.1949	0.1999
c <sub>5</sub>	1	0.2204	0.2276	0.2095
c <sub>6</sub>	1	0.3846	0.3959	0.2324
c <sub>7</sub>	2	0.2130	0.0502	0.1115
c <sub>8</sub>	2	0.1248	0.0794	0.1104
c <sub>9</sub>	2	0.1081	0.0954	0.1079
c <sub>10</sub>	2	0.1248	0.1259	0.1104
c <sub>11</sub>	2	0.2130	0.2254	0.1115
c <sub>12</sub>	3	0.1328	0.0224	0.0609
c <sub>13</sub>	3	0.0917	0.0441	0.0675
c <sub>14</sub>	3	0.0917	0.0712	0.0675
c <sub>15</sub>	3	0.1328	0.1323	0.0609
c <sub>16</sub>	4	0.1035	0.0126	0.0378
c <sub>17</sub>	4	0.0891	0.0348	0.0444
c <sub>18</sub>	4	0.1035	0.0758	0.0378
c <sub>19</sub>	5	0.1014	0.0100	0.0247
c <sub>20</sub>	5	0.1014	0.0366	0.0247
c <sub>21</sub>	6	0.1132	0.0103	0.0135

$(4n+2)$ -site conjugated ones with positive CREs. These circuits are the origin of high aromaticity in these hydrocarbons. In contrast, all the circuits in polyacenequinododimethides **7–18** are  $(4n+2)$ -site nonconjugated ones, so are still more or less aromatic with positive CREs. According to the extended Hückel rule proposed by Hosoya et al.,<sup>[35,36]</sup>  $(4n+2)$ -site conjugated and nonconjugated circuits contribute to aromaticity. In general, larger circuits contribute less to aromaticity than smaller ones. For polyacene molecules, outer circuits have larger CREs as long as they have the same area. However, a larger number of circuits contribute to the local aromaticity of inner rings. For polyacenequinododimethide molecules, circuits farther from the two methylene groups have larger CREs, if the areas are the same.

The total  $\pi$ -current-density maps for **1–18** are presented in Figure 3, in which counterclockwise currents indicate diatropicity. For the polyacene series, a strong diatropic perimeter current is observed, which is rather concentrated on the inner benzene rings.<sup>[29,37]</sup> A larger number of diatropic circuit currents are superposed at the inner rings. As pointed out by Havenith et al.,<sup>[30]</sup> a similar behavior of  $\alpha,\omega$ -dicyclopenta-fused polyacenes indicates that these molecules behave like polyacenes with  $4n+2$   $\pi$  electrons, although the only conjugated circuit in these hydrocarbons is a peripheral  $4n$ -site one. Polyacenequinododimethides are similar in this sense to  $\alpha,\omega$ -dicyclopenta-fused polyacenes. It seems quite likely that a molecule with a large polyacene moiety behaves like a polyacene whatever the remaining part of the molecule might be.

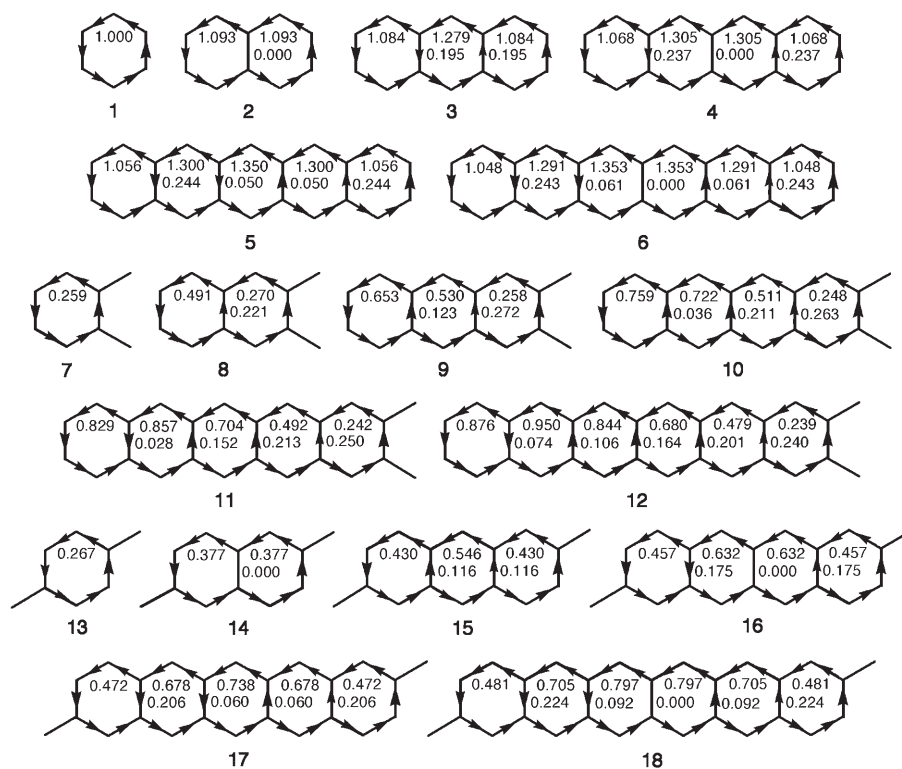


Figure 3. Maps of total  $\pi$ -current density for **1–18**.

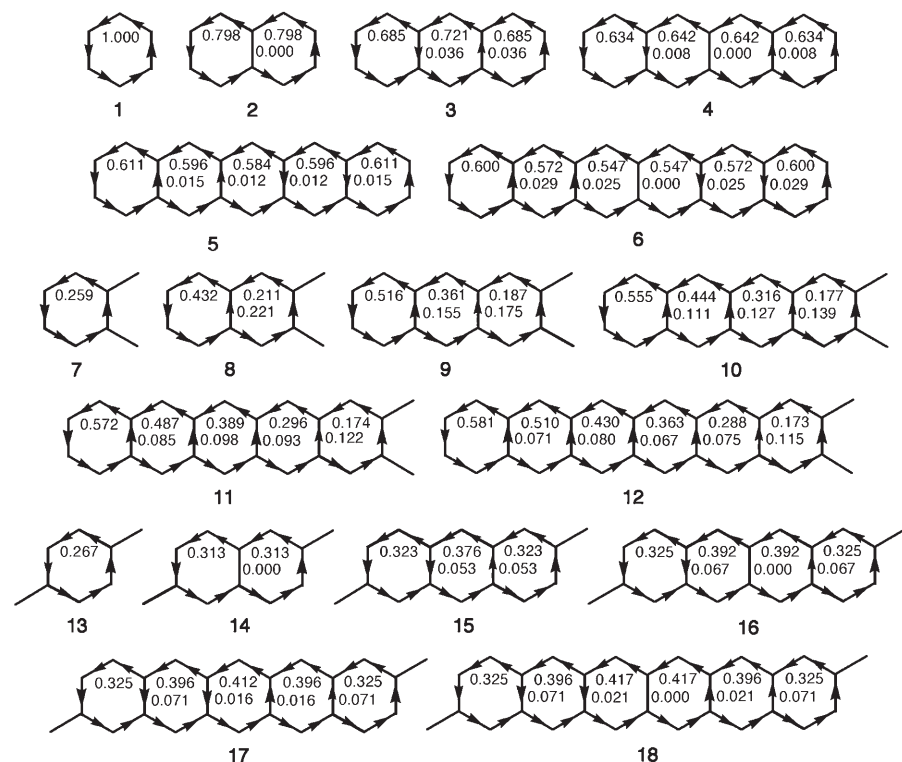


Figure 4. Maps of geometry-independent  $\pi$ -current density for **1–18**.

The maps of geometry-independent  $\pi$ -current density for **1–18** are shown in Figure 4. Those for **1–6** are the same as

reported in reference [21]. Figure 5 gives the plot of the intensity of a geometry-independent  $\pi$  current against the BRE for all peripheral  $\pi$  bonds in **1–18**. An excellent corre-

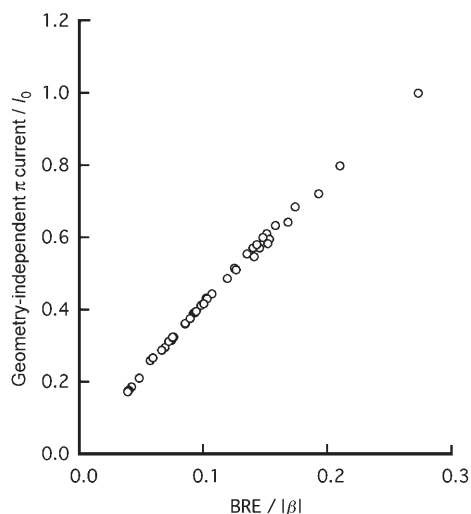


Figure 5. Correlation between the intensity of the geometry-independent  $\pi$  current and BRE for all peripheral  $\pi$  bonds in **1–18**.

lation was found between the two quantities with a correlation coefficient of 0.998, which clearly indicates that not only the BRE for a peripheral  $\pi$  bond but also the intensity of a geometry-independent  $\pi$  current can be used as an indicator of local aromaticity.<sup>[19]</sup> As may be inferred from [Eq. (5)], the approximate value for the BRE is obtained by dividing the intensity of the corresponding geometry-independent  $\pi$  current by 4.5. Notably, the BRE is associated with the stabilization energy due to all circuits that share the  $\pi$  bond concerned.<sup>[16–19]</sup> Therefore, the discussion of local aromaticity made with the BREs can be applied to the geometry-independent  $\pi$  currents.

Finally, the circuit-current diamagnetic susceptibilities, that is, the circuit contributions to the ring-current susceptibilities, calculated for hexacene (**6**), hexacene-2,3-quinododimethide (**12**), and hexacene-2,10-quinododimethide (**18**) are presented in Table 5. Notably, for all these species, large circuits with small CREs contribute much to the ring-current diamagnetic susceptibility, because the circuit-current susceptibility is proportional to the square of the circuit area. This is the primary reason why the ring-current diamagnetic susceptibility and diamagnetic susceptibility exaltation are not reliable indices of aromaticity. They overestimate the aromaticity of large but less-aromatic circuits.

## Conclusions

Polyacenequinododimethides are more or less aromatic and diatropic even if they have no aromatic conjugated circuits. Such an apparent dichotomy with respect to aromatic character was interpreted with our graph theory of aromaticity

Table 5. Circuit-current diamagnetic susceptibilities for hexacene (**6**) and hexacenequinododimethides **12** and **18**.

Circuit	Area [ $S_0$ ]	Circuit-current diamagnetic susceptibilities [ $\chi_0$ ]		
		<b>6</b>	<b>12</b>	<b>18</b>
c <sub>1</sub>	1	0.3846	0.1331	0.2324
c <sub>2</sub>	1	0.2204	0.1780	0.2095
c <sub>3</sub>	1	0.1916	0.1874	0.1999
c <sub>4</sub>	1	0.1916	0.1949	0.1999
c <sub>5</sub>	1	0.2204	0.2276	0.2095
c <sub>6</sub>	1	0.3846	0.3959	0.2324
c <sub>7</sub>	2	0.4260	0.1005	0.2231
c <sub>8</sub>	2	0.2496	0.1589	0.2208
c <sub>9</sub>	2	0.2163	0.1907	0.2159
c <sub>10</sub>	2	0.2496	0.2518	0.2208
c <sub>11</sub>	2	0.2496	0.4507	0.2231
c <sub>12</sub>	3	0.3983	0.0672	0.1827
c <sub>13</sub>	3	0.2750	0.1324	0.2025
c <sub>14</sub>	3	0.2750	0.2135	0.2025
c <sub>15</sub>	3	0.3983	0.3969	0.1827
c <sub>16</sub>	4	0.4139	0.0506	0.1514
c <sub>17</sub>	4	0.3563	0.1391	0.1774
c <sub>18</sub>	4	0.4139	0.3031	0.1514
c <sub>19</sub>	5	0.5072	0.0499	0.1234
c <sub>20</sub>	5	0.5072	0.1831	0.1234
c <sub>21</sub>	6	0.6790	0.0618	0.0807

and magnetotropy. Many nonconjugated circuits were found to contribute collectively to aromaticity and diatropicity. As a result, at least, benzene rings far from the *exo*-methylenes exhibit considerable local aromaticity. This fact strongly suggests that the conjugated circuit model<sup>[1,2]</sup> might not be applicable to semibenzenoid hydrocarbons without conjugated circuits. It is also clear that the quinododimethides cannot be used as polyene references in estimating the ASEs of structurally related species. The aromaticity and diatropicity of fulminene-2,10-quinododimethide and its homologues<sup>[7,8]</sup> can be interpreted in essentially the same manner. Work in this direction is in progress.

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