CHEMICAL GRAPH THEORY OF FIBONACENES

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Abstract

Fibonacenes (zig-zag unbranched catacondensed benzenoid hydrocarbons) are a class of polycyclic conjugated systems whose molecular graphs possess remarkable properties, often related with the Fibonacci numbers. This article is a review of the chemical graph theory of fibonacenes, with emphasis on their Kekulé–structure–related and Clar–structure–related properties.

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0. FIBONACCI NUMBERS

The sequence of integers $F_0, F_1, F_2, F_3, \ldots$, named after Leonardo Pisano aka Fibonacci (1170–1250), is defined by means of the recurrence relation

$$F_n = F_{n-1} + F_{n-2}$$

and by means of the initial conditions

$$F_0 = 0 \quad ; \quad F_1 = 1 \ .$$

Thus, $F_2 = 1$, $F_3 = 2$, $F_4 = 3$, $F_5 = 5$, $F_6 = 8$, $F_7 = 13$, $F_8 = 21$, $F_9 = 34$, $F_{10} = 55$, etc.

The mathematical theory of Fibonacci numbers is very interesting and can be found in pertinent books (for instance, in [1, 2]) or in the articles published in the journal "*Fibonacci Quarterly*".[1]

1. INTRODUCTION

Fibonacenes are unbranched catacondensed benzenoid hydrocarbons in which all non-terminal hexagons are angularly annelated. Their structure should be evident already from the examples depicted in Fig. 1.

The name *fibonacene*[2] was proposed by Balaban in 1989 [3], although it seems to be first mentioned somewhat earlier [4]. The name is due to the (long known [5, 6, 7]) fact that the Kekulé structure count of fibonacenes coincides with the Fibonacci numbers; for details see below.

In order to define fibonacenes (or more precisely: the molecular graphs of fibonacenes) in a more precise manner, we need to recall some basic notions from the theory of benzenoid systems [8]. In what follows we employ the terminology and definitions employed in an earlier review [9].

[1] When reading a book or an article in which Fibonacci numbers are encountered, one should always check the way in which these are defined. Some authors use the initial conditions $F_0 = F_1 = 1$, and thus their "Fibonacci numbers" are shifted by one relative to the present ones.

[2] In view of some recent disputes concerning the spelling of the name, it may be useful to repeat Balaban's original words from [3]: *We are aware that fibonaccenes would be etymologically more suitable but we suppressed one c for the simplicity and for similarity with the established name "acenes".*

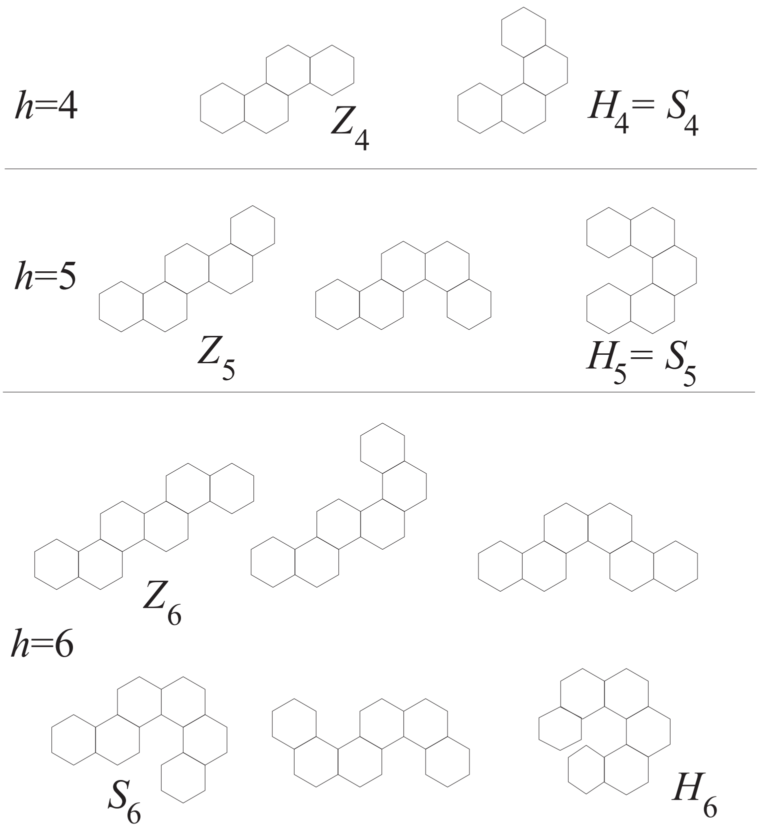

Figure 1: The fibonacenes with four, five, and six hexagons; cf. Fig. 3.

Definition 1. A *hexagonal system* is a connected plane graph without cut-vertices in which all inner faces are hexagons (and all hexagons are faces), such that two hexagons are either disjoint or have exactly one common edge, and no three hexagons share a common edge.

Definition 2. A hexagonal system is said to be *simple* if it can be embedded into the regular hexagonal lattice in the plane without overlapping of its vertices. Hexagonal systems that are not simple are called *jammed*.

Hexagons sharing a common edge are said to be *adjacent* or *neighboring*. Two hexagons of a hexagonal system may have either two common vertices (if they are adjacent) or none (if they are not adjacent). A vertex of a hexagonal system belongs to at most three hexagons. A vertex shared by three hexagons is called an *internal vertex* of the respective hexagonal system.

Definition 3. A hexagonal system is said to be *catacondensed* if it does not possess internal vertices. A hexagonal system is said to be *pericondensed* if it possesses at least one internal vertex.

A hexagon of a catacondensed hexagonal system has either one, two, or three neighboring hexagons. A hexagon having exactly one neighboring hexagon is said to be *terminal*. A hexagon having three neighboring hexagons is said to be *branched*.

Definition 4. A catacondensed hexagonal system possessing at least one branched hexagon is said to be a *branched catacondensed hexagonal system*. A catacondensed hexagonal system without branched hexagons is called a *hexagonal chain*.

A hexagonal chain with h hexagons, $h \geq 2$, possesses two terminal hexagons and $h-2$ hexagons that have two neighbors. Hexagons being adjacent to exactly two other hexagons are classified as angularly or linearly annelated. A hexagon adjacent to exactly two other hexagons possesses two vertices of degree 2. If these two vertices are adjacent, then the hexagon is *angularly annelated*, if these two vertices are not adjacent, then it is *linearly annelated*.

Definition 5. A *fibonacene* is a hexagonal chain without linearly annelated hexagons.

One should note that fibonacenes may be either simple or jammed. Among the examples shown in Fig. 1 only the last one (with $h = 6$) is jammed; it corresponds to the chemical compound called “hexahelicene”. In many graph–theory–based studies (including those outlined in the present paper) it is convenient to consider the class of all fibonacenes, including both simple and jammed. Sometimes, however, jammed systems would be excluded, see e. g. [8]. Anyway, all properties of fibonacenes discussed in this review are independent of whether these are or are not simple.

Throughout this article h always denotes the number of hexagons.

In what follows, for the sake of brevity, a hexagonal chain and a fibonacene with h hexagons will be referred to as an h-hexagonal chain and an h-fibonacene, respectively.

2. SYMMETRY AND ENUMERATION OF FIBONACENES

According to symmetry, fibonacenes can be mirror–symmetric (S), centro–symmetric (C) and asymmetric (A). Characteristic examples of these symmetry types are shown in Fig. 2.

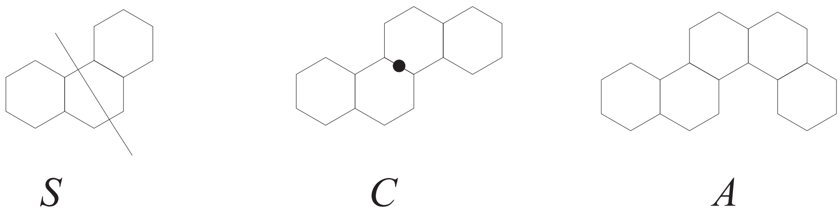

Figure 2: Examples (the smallest possible) of a mirror–symmetric (S), centro–symmetric (C) and asymmetric (A) fibonacenes. The symmetry axis and the center of symmetry are indicated in S and C, respectively.

If $S(h)$, $C(h)$, and $A(h)$ are, respectively, the number of mirror–symmetric, centro–symmetric, and asymmetric h-fibonacenes, then for $h \geq 3$ [3],

$$S(h) = \begin{cases} 2^{h/2-2} & \text{if } h \text{ is even} \\ 2^{(h-1)/2-1} & \text{if } h \text{ is odd} \end{cases}$$

$$C(h) = \begin{cases} 2^{h/2-2} & \text{if } h \text{ is even} \\ 0 & \text{if } h \text{ is odd} \end{cases}$$

$$A(h) = \begin{cases} 2^{h-4} - 2^{h/2-2} & \text{if } h \text{ is even} \\ 2^{h-4} - 2^{(h-1)/2-2} & \text{if } h \text{ is odd} \end{cases} .$$

The total number of h-fibonacenes is $T(h) = S(h) + C(h) + A(h)$, and thus

$$T(h) = \begin{cases} 2^{h-4} + 2^{h/2-2} & \text{if } h \text{ is even} \\ 2^{h-4} + 2^{(h-1)/2-2} & \text{if } h \text{ is odd} \end{cases} . \tag{1}$$

It is easy to see that

$$\lim_{h\to\infty} \frac{A(h)}{T(h)} = 1$$

implying that almost all fibonacenes are asymmetric. (Analogous results are known also for hexagonal systems in general, as well as for catacondensed and pericondensed hexagonal systems [10].)

Denote by $J(h)$ the number of jammed h-fibonacenes. The $J(h)$-values were determined by Balaban for the first few values of h [3]:

h	3	4	5	6	7	8	9	10
$J(h)$	0	0	0	1	2	5	11	26
$T(h)$	1	2	3	6	10	20	36	72

Although at the first glance, the $J(h)$-values appear to be much smaller than $T(h)$, in the limit $h \to \infty$, almost all h-fibonacenes happen to be jammed.

3. BASIC PROPERTIES OF FIBONACENES

A molecular graph representing any h-fibonacene has $4h + 2$ vertices, of which $2h + 4$ are of degree 2 and $2h - 2$ of degree 3. This graph has $5h + 1$ edges, of which $h + 4$ connect two vertices of degree 2, $2h$ connect two vertices of degree 3, and $2h - 3$ connect vertices of degree 2 and 3. For more detail on these "anatomic" properties see [8].

The Wiener index of hexagonal systems was much investigated [9], among which also of fibonacenes. Among the fibonacenes with a fixed number of hexagons three are extremal with regard to their Wiener indices: the helicene (H_h), the zig-zag fibonacene (Z_h), and the serpent (S_h), see Fig. 3. Their Wiener indices conform to the expressions:

$$W(H_h) = (8\,h^3 + 72\,h^2 - 26\,h + 27)/3 \tag{2}$$

$$W(Z_h) = (16\,h^3 + 24\,h^2 + 62\,h - 21)/3 \tag{3}$$

$$W(S_h) = \begin{cases} (32\,h^3 + 168\,h^2 - 6\,h + 81)/9 & \text{if } h = 3k\ ,\ k = 1,2,3,\ldots \\ (32\,h^3 + 168\,h^2 - 6\,h + 49)/9 & \text{if } h = 3k+1\ ,\ k = 0,1,2,\ldots \\ (32\,h^3 + 168\,h^2 - 6\,h + 16)/9 & \text{if } h = 3k+2\ ,\ k = 0,1,2,\ldots \end{cases} \tag{4}$$

Formulas (2), (3), and (4) seem to have been first reported in [11], [11], and [12], respectively. It has been shown [13] that H_h has the smallest Wiener index among all h-hexagonal chains, whereas S_h has the smallest Wiener index among all simple h-hexagonal chains [12]. The system Z_h has the greatest Wiener index among all h-fibonacenes.

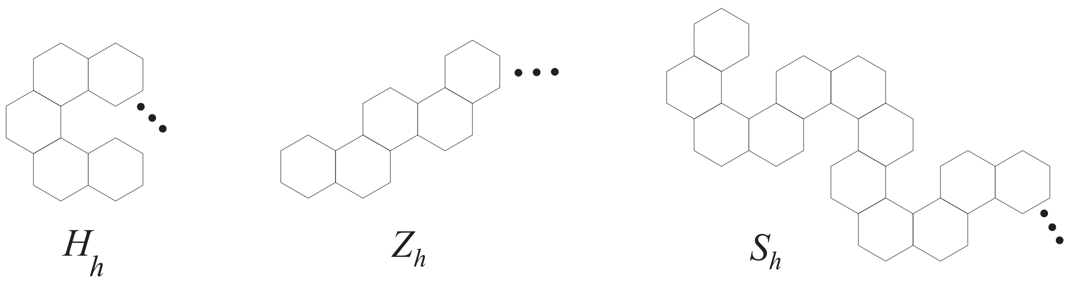

Figure 3: Three extremal fibonacenes: helicene (H_h), zig-zag fibonacene (Z_h), and serpent (S_h); see also Fig. 1.

The same fibonacene Z_h was shown to have maximal energy among all h-hexagonal chains [14].

A peculiar result was obtained by considering the average Wiener index $W_{avr}(h)$ in the set $\mathcal{F}(h)$ of all fibonacenes with h hexagons. By definition,

$$W_{avr}(h) = \frac{1}{|\mathcal{F}(h)|} \sum_{G \in \mathcal{F}(h)} W(G) \ .$$

It was shown [15] that $W_{avr}(h)$ is necessarily an integer, implying that the sum of the Wiener indices of all h-fibonacenes is divisible by the number $T(h)$ of h-fibonacenes. (Recall that $T(h)$ is given by Eq. (1).) Furthermore,

$$W_{avr}(h) = 4\,h^3 + 16\,h^2 + 6\,h + 1 \ .$$

4. KEKULÉ STRUCTURES OF FIBONACENES

Denote by $K\{H\}$ the number of Kekulé structures (or, in the language of graph theory, the number of perfect matchings) of the hexagonal system H .

Before formulating the main result of this section, we state without proof a more general regularity for the number of Kekulé structures.

Let R be a (molecular) graph and let u and v be its two adjacent vertices. Let S be another (molecular) graph and x and y its adjacent vertices. Let the graphs H_1, H_2, H_3, H_4

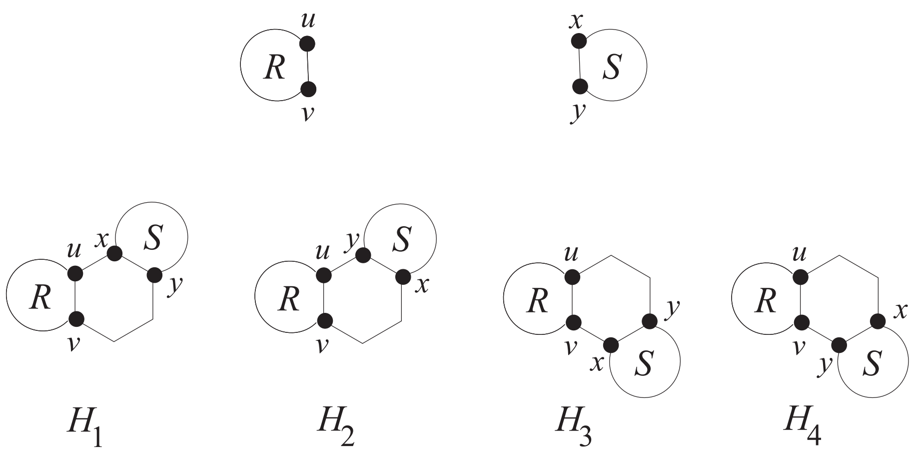

Figure 4: Four different ways in which angular annelation can be done.

be obtained by annelating (in an angular manner) the fragments R and S to a hexagon, as shown in Fig. 4.

Theorem 1. (a) If the fragments R and S have both an even number of vertices, then $K\{H_1\} = K\{H_2\} = K\{H_3\} = K\{H_4\}$.
(b) If the fragments R and S have both an odd number of vertices, then $K\{H_1\} = K\{H_4\}$ and $K\{H_2\} = K\{H_3\}$, but $K\{H_1\}$ and $K\{H_2\}$ may differ.
(c) If the fragment R has even and S odd number of vertices (or vice versa), then $K\{H_1\} = K\{H_2\} = K\{H_3\} = K\{H_4\} = 0$.

It is immediate to see that part (a) of Theorem 1 is directly applicable to fibonacenes, implying:

Corollary 1.1. All h-fibonacenes have equal number of Kekulé structures.

A long–time known [5, 6], easy–to–prove, and frequently used [7, 8, 17, 18, 19, 20, 21, 22], result on Kekulé structures is the following:

Theorem 2. The number of Kekulé structures of the zig-zag fibonacene Z_h is equal to the $(h+2)$-th Fibonacci number,

$$K\{Z_h\} = F_{h+2} \quad , \quad h \geq 1 \; .$$

Corollary 2.1. The number of Kekulé structures of any h-fibonacene is equal to the

$(h+2)$-th Fibonacci number.

5. CLAR STRUCTURES OF FIBONACENES

Denote the number of Clar structures [8, 16] of a hexagonal system H by $C(H)$, and its Clar number, that is the number of aromatic sextets in any of the Clar structures, by $Cl(H)$. Recall that the calculation of both $C(H)$ and $Cl(H)$ is not an easy task, and was the subject of several earlier studies [23, 24, 25].

In connection with the Clar structures one can establish results similar to those for Kekulé structures (but not involving Fibonacci numbers). First we have an analog of Theorem 1:

Theorem 3. Using the notation employed in Theorem 1 (cf. Fig. 4), the following holds.
(a) If the fragments R and S have both an even number of vertices, then $C\{H_1\} = C\{H_2\} = C\{H_3\} = C\{H_4\}$ and $Cl\{H_1\} = Cl\{H_2\} = Cl\{H_3\} = Cl\{H_4\}$.
(b) If the fragments R and S have both an odd number of vertices, then $C\{H_1\} = C\{H_4\}$, $C\{H_2\} = C\{H_3\}$, and $Cl\{H_1\} = Cl\{H_4\}$, $Cl\{H_2\} = Cl\{H_3\}$, but $C\{H_1\}$ and $C\{H_2\}$ as well as $Cl\{H_1\}$ and $Cl\{H_2\}$ may differ.
(c) If the fragment R has even and S odd number of vertices (or vice versa), then $C\{H_1\} = C\{H_2\} = C\{H_3\} = C\{H_4\} = 0$ and $Cl\{H_1\} = Cl\{H_2\} = Cl\{H_3\} = Cl\{H_4\} = 0$.

Corollary 3.1. All h-fibonacenes have equal number of Clar structures and equal Clar numbers.

In Fig. 5 are shown the Clar structures of the zig-zag h-fibonacenes for $h = 4$ and $h = 5$. By inspection of this figure one easily reaches the following general conclusion:

Theorem 4. The zig-zag fibonacene Z_h has $\lceil h/2 \rceil$ aromatic sextets. If h is odd, it has a unique Clar structure. If h is even, it has $h/2 + 1$ distinct Clar structures.

Corollary 4.1. The number of Clar structures of any h-fibonacene is equal to 1 (if h is odd) or to $h/2 + 1$ (if h is even). The Clar number of any h-fibonacene is $\lceil h/2 \rceil$.

The fact that the modes of cyclic conjugation and the distribution of π-electrons, as represented by the Clar structures, are significantly different in fibonacenes with odd and

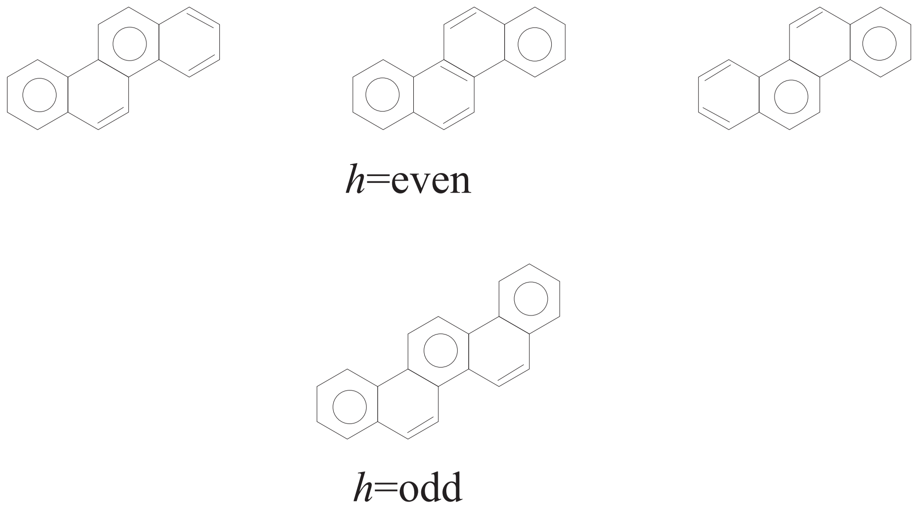

Figure 5: Clar structures of the zig-zag 4– and 5-fibonacenes. Here $C(Z_4) = 3$, $Cl(Z_4) = 2$ and $C(Z_5) = 1$, $Cl(Z_5) = 3$.

even number of hexagons attracted some attention [26, 27, 28]. However, no significant difference between the stability and "aromaticity" of odd and even fibonacenes could be envisaged by any of the theoretical approaches, in harmony with the existing experimental data [29, 30].

6. FIBONACENES AS MAXIMUM CYCLE RESONANT HEXAGONAL CHAINS

In this section we show that fibonacenes can be characterized among hexagonal chains using a concept from the resonance theory. To state and prove the result, some preparation is needed.

Disjoint cycles of a graph G are called *mutually resonant* if there exists a Kekulé structure M of G such that any of the given cycles in M-alternating. G is *k-cycle resonant* if it contains at least k disjoint cycles and any r disjoint cycles, where $1 \leq r \leq k$, are mutually resonant.

Let H be a hexagonal system and let P be a path whose endvertices are of degree three in H and all the other vertices are of degree two in H. Then we say that P is a *3,3-boundary path* of H. Then we have the following nice characterization of 2-cycle resonant hexagonal systems proved by Gou and Zhang in [31].

Theorem 5. Let H be a hexagonal system with at least two disjoint hexagons. Then H is 2-cycle resonant if and only if H is a catacondensed hexagonal system with no 3,3-boundary path of even length.

In fact, 2-cycle resonance is critical when considering higher cycle resonance, as the next result from [31] asserts.

Theorem 6. Let H be a 2-cycle resonant hexagonal system and let k be the maximum number of disjoint cycles in H. Then H is k-cycle resonant.

Combining Theorems 5 and 6 we obtain the following result that characterizes fibonacenes among hexagonal chains. Call a hexagonal system H *maximum cycle resonant* if H is k-cycle resonant, where k is the maximum number of disjoint cycles of H. Then:

Theorem 7. Let H be a hexagonal chain. Then H is a fibonacene if and only if H is maximum cycle resonant.

Proof. If a hexagonal chain H consists of one or two hexagons, the theorem is clear. Suppose H contains at least three hexagons. Then we easily see that H contains no 3,3-boundary path of even length if and only if H is a fibonacene. Then Theorem 5 implies that H is 2-cycle resonant if and only if H is a fibonacene and Theorem 6 completes the argument. □

Theorem 7 has been first stated explicitly by Shiu, Lam, and Zhang in [32]. Their somewhat more complicated proof also uses results of [31]. In [32] two extremal properties concerning the number of matchings and the number of independent sets are also proved for fibonacenes. We also add that in [33] the theory of cycle resonance is further developed. In particular, an algorithm is proposed that determines whether a planar 2-connected bipartite graph is 1-cycle resonant.

7. FIBONACENES AS FIBONACCI CUBES

We continue with the role of fibonacenes in the resonance theory. In this section we demonstrate a surprising connection between fibonacenes and graphs that were introduced in theoretical computer science as a model for interconnection networks.

Let H be a hexagonal system. Then the vertex set of the *resonance graph* $R(H)$ of H consists of all Kekulé structures of H, where two Kekulé structures are adjacent whenever their symmetric difference is the edge set of a hexagon of H; an example of a fibonacene and its resonance graph is shown on Fig. 6.

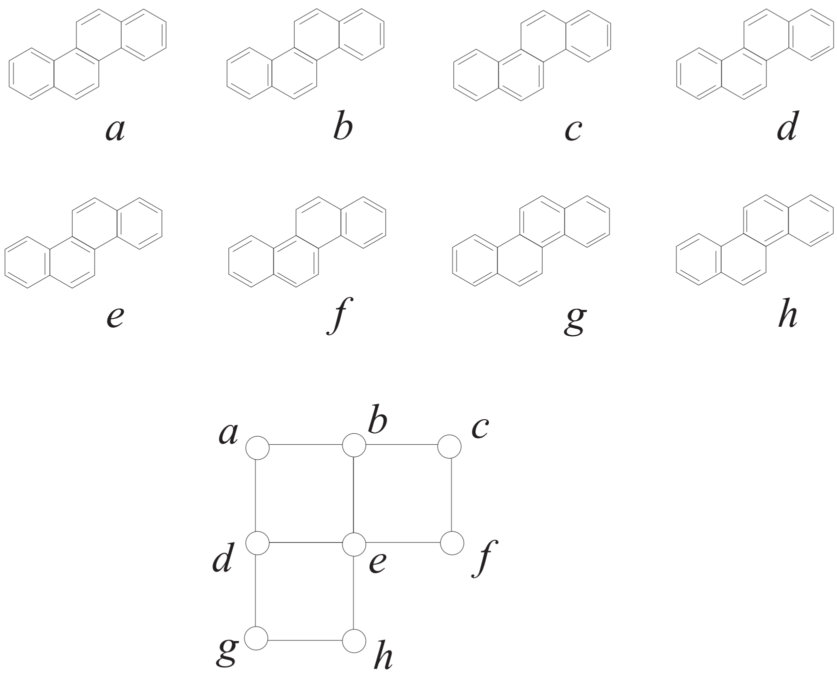

Figure 6: The Kekulé structures of Z_4 and the resonance graph $R(Z_4)$.

Resonance graphs have been independently introduced in the chemical literature [34, 35, 36] as well as in the mathematical literature [37]. For more result on the resonance graphs see [38, 39, 40, 41, 42].

Consider now the following seemingly unrelated concept that has been introduced in [43, 44] as a model for interconnection networks, see also [45, 46, 47, 48].

The vertex set of the *Fibonacci cube* Γ_n, $n \geq 1$, is the set of all binary strings $b_1 b_2 \ldots b_n$ containing no two consecutive ones. Two vertices are adjacent in Γ_n if they differ in precisely one bit, cf. Fig. 7 where the first four Fibonacci cubes are shown.

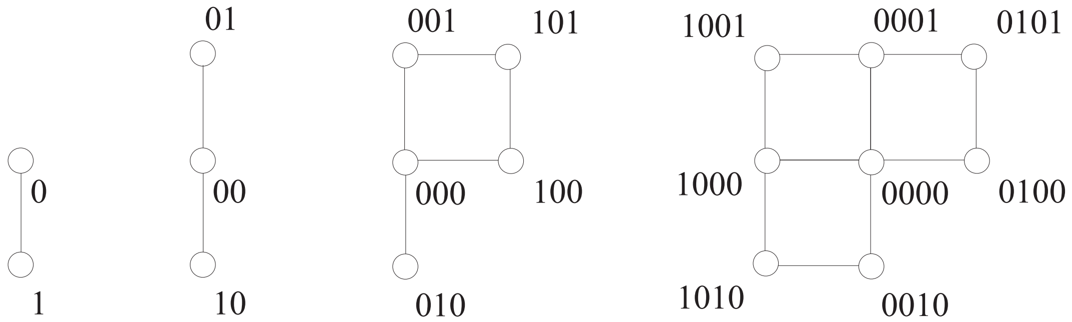

Figure 7: The Fibonacci cubes Γ_1, Γ_2, Γ_3, and Γ_4.

Compare Figures 6 and 7 to note that Γ_4 is just the resonance graph of Z_4, that is, $\Gamma_4 = R(Z_4)$. This is not a coincidence, as our final theorem from [49] asserts.

Theorem 8. Let H be an arbitrary fibonacene with n hexagons. Then $R(H) = \Gamma_n$.

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