Clar Covers of Overlapping Benzenoids: Case of Two Identically-Oriented Parallelograms

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Abstract: We present a complete set of closed-form formulas for the ZZ polynomials of five classes of composite Kekuléan benzenoids that can be obtained by overlapping two parallelograms: generalized ribbons $Rb$, parallelograms $M$, vertically overlapping parallelograms $M\oplus M$, horizontally overlapping parallelograms $M\otimes M$, and intersecting parallelograms $M\times M$. All formulas have the form of multiple sums over binomial coefficients. Three of the formulas are given with a proof based on the interface theory of benzenoids, while the remaining two formulas are presented as conjectures verified via extensive numerical tests. Both of the conjectured formulas have the form of a $2 \times 2$ determinant bearing close structural resemblance to analogous formulas for the number of Kekulé structures derived from the John-Sachs theory of Kekulé structures.

Keywords: enumeration of Clar covers; ZZ polynomials; Clar covering polynomials; parallelogram-shaped benzenoids

1. Introduction

Benzenoid hydrocarbons are an intensively studied class of aromatic molecules attracting attention of scientists specializing in various fields of chemistry including organic synthesis [1,2] and characterization [3–5], combustion chemistry [6,7], nanoscience [8,9], atmospheric chemistry [10,11], astrochemistry [12,13], computational chemistry [14,15], and chemical graph theory [16,17]. Enumeration and construction of Clar covers for benzenoid hydrocarbons constitutes one of the important branches of chemical graph theory. From the point of view of a chemist, a Clar cover is a generalized resonance structure that can be constructed for a given benzenoid $B$, in which the tetravalent character of each carbon atom is ensured by making it participate either in a two-center $\pi$ bond with one of its closest-neighbor carbon atoms or in a delocalized aromatic $\pi$ sextet with the other five carbon atoms in the same benzene ring. From the point of view of a mathematician, the benzenoid $B$ is a finite plane graph embedded in a hexagonal lattice [16] and a Clar cover is a spanning subgraph of $B$ whose components are either $K_2$ or $C_6$ ($K_2$ denotes a complete graph on two vertices and $C_6$ denotes a cycle graph of length six). To unify these two points of view, we establish the following correspondences to be used whenever convenient: a vertex in $B$ $\equiv$ a carbon atom in $B$, an edge in $B$ $\equiv$ a bond in $B$, the edge in $K_2$ $\equiv$ a double bond, $C_6$ $\equiv$ an aromatic ring, and an edge in $C_6$ $\equiv$ an aromatic bond.

In general, a large number of Clar covers can be constructed for every Kekuléan benzenoid $B$. Finding this number—usually denoted by $C$ and referred to as the Clar count—can be quite a complex task, particularly for large benzenoids of irregular shapes. The main goal of the current report is to show that for a large group of complex-shape benzenoids obtained by overlapping two identically
oriented parallelograms, $M(m_1, n_1)$ and $M(m_2, n_2)$, the number $C$ can be readily computed from the structural parameters $m_1$, $n_1$, $m_2$, and $n_2$ of the two parallelograms $M(m_1, n_1)$ and $M(m_2, n_2)$ and the structural parameters $\mu$ and $\nu$ of the parallelogram-shaped overlapping region $M(\mu, \nu)$. To derive these expressions, we need to introduce several auxiliary concepts.

2. Counting Clar Covers

The set of Clar covers can be naturally partitioned with respect to their order, i.e., the number of the aromatic rings $C_6$ used to construct a given Clar cover. Each such subset of Clar covers containing exactly $k$ aromatic rings is referred to as the Clar covers of order $k$ and the cardinality of this subset is denoted as $c_k$. The set of Clar covers of order 0 is identical to the set of Kekulé structures of $B$, giving the identification between $c_0$ and the Kekulé count $K$. The finite nature of $B$ (consisting of $n$ carbon atoms) equips us with a natural upper bound $\left\lceil \frac{n}{2} \right\rceil$ for the maximal number of aromatic sextets $C_6$—usually referred to as the Clar number $Cl$—that can be placed in $B$. Consequently, the number of subsets in the order partitioning is also finite and their cardinalities can be expressed by the sequence $(c_0, c_1, \ldots, c_{Cl})$. A generating function of this sequence

$$ZZ(B, x) = \sum_{k=0}^{Cl} c_k x^k$$

is usually referred to as the Clar covering polynomial or as the Zhang-Zhang (ZZ) polynomial, after the names of two Chinese mathematicians, Fuji Zhang and Heping Zhang, who first introduced it to the field of chemical graph theory about 25 years ago [18–21]. The ZZ polynomial of $B$ decodes its most important topological invariants:

- Kekulé count $K = c_0 = ZZ(B, 0)$,
- Clar count $C = c_0 + \ldots + c_{Cl} = ZZ(B, 1)$,
- Clar number $Cl = \text{degree}(ZZ(B, x))$,
- the number of Clar formulas $c_{Cl}$,
- and the first Herndon number $h_1 = c_1$ [22].

The most obvious way to compute the ZZ polynomial of $B$ is to construct the full set of Clar covers of $B$ and simply count the Clar covers of each order. This is most conveniently performed by a recursive assignment of single, double, and aromatic coverings to selected edges of $B$ until the covering character of all edges in $B$ is determined. An example of such a procedure is given in Figure 1, where the set of 25 Clar covers of the parallelogram $M(2, 3)$ is generated in a recursive manner by forming a tree, in which the root corresponds to the original structure $M(2, 3)$ and every leaf to a distinct Clar cover of $M(2, 3)$. Since the number of Clar covers of order 0, 1 and 2 is 10, 12, and 3, respectively, the resulting ZZ polynomial is given by $ZZ(M(2, 3), x) = 10 + 12x + 3x^2$. Unfortunately, this approach is feasible but for the smallest benzenoids. For $M(10, 10)$ with 240 carbon atoms, the number of Clar covers comprises more than 8 millions resonance structures and it is completely impractical to represent it graphically in any way. Fortunately, ZZ polynomials possess attractive recurrence properties (see Theorems 3–6 of [18] or Properties 1–7 of [23]) allowing computing them in a recursive manner. These properties made it possible to use a recursive algorithm [18,23,24], which performs consecutive decompositions of $B$ to compute the final ZZ polynomial recursively from the ZZ polynomials of the nodes of the decomposition tree. Our group designed an automatized computer program called ZZ Decomposer [25–27], which can be used first for a graphical definition of the studied benzenoid by selecting a subset of cells in a hexagonal lattice and then for computation or derivation of its ZZ polynomial. Application of ZZ Decomposer to $M(10, 10)$ yields the desired result within a few seconds. (For details, see Figure 2) Another exemplary application of ZZ Decomposer is the otherwise very tedious determination of ZZ polynomials of all the isomers of carbon fullerenes also presented in the recent volume of “Symmetry” [28].
Figure 1. 25 Clar covers of the parallelogram $M(2, 3)$ can be constructed by a recursive assignment of single (no covering, symbol S), double ($K_2$ covering, symbol D), or aromatic ring ($C_6$ covering, symbol R) character to the edges of $M(2, 3)$ marked with a red circle. Each Clar cover of order 0 (i.e., Kekulé structure, depicted in cyan frames) contributes a term 1 to $\text{ZZ}(M(2, 3), x)$, each Clar cover of order 1 (purple frames) contributes a term $x$ to $\text{ZZ}(M(2, 3), x)$, and each Clar cover of order 2 (orange frames) contributes a term $x^2$ to $\text{ZZ}(M(2, 3), x)$. The resulting ZZ polynomial of $M(2, 3)$ is thus equal to $10 + 12x + 3x^2$.

Figure 2. Brute-force computation of the ZZ polynomial of the $M(10, 10)$ parallelogram using ZZDecomposer takes about 14 s on a desktop computer.

More importantly, ZZDecomposer can be used for discovering closed-form formulas for ZZ polynomials of whole families of isostructural benzenoids. First such formulas were discovered without ZZDecomposer by Zhang and Zhang [18–20] in their seminal papers for the simplest families of benzenoids: polyacenes $L(n)$ [19], hexagonal chains [19], branched catacondensed benzenoids and catacondensed ladders [19], catacondensed all-benzenoids [19], and prolate rectangles [21]. Soon, other developments along this line appeared allowing finding closed-form formulas for other
simple benzenoid structures: 2- and 3-tier benzenoid strips [29], one family of oblate rectangles [24],
cyclo-polyphenacenes [30,31], and parallelograms [32]. In particular, the work performed by Gutman
and Borovičanin [32,33] permits us to write down the formula for the ZZ polynomial of the
parallelogram $M(m, n)$ with arbitrarily large values of the parameters $m$ and $n$ as

$$
ZZ(M(m, n), x) = \sum_{k=0}^{\min(m, n)} \binom{m}{k} (\frac{n}{m} + n - k)x^k
$$

(2)

$$
= \sum_{k=0}^{\min(m, n)} \binom{m}{k} (\frac{n}{m})(1 + x)^k
$$

(3)

However, it was really the development of ZZDecomposer [25–27] which made such
discoveries possible even for complicated structures [23,25,34,35] including generalized oblate
rectangles [36], 3-, 4-, and 5-tier regular benzenoid strips [37,38], chevrons and generalized
chevrons [33], zigzag-edge coronoids and fenestrenes [39], multiple zigzag chains [40], hexagons
with corner defects [41], and ribbons [42]. At the moment, the only two remaining families of
basic benzenoids for which a closed-form ZZ polynomial formula is not available are hexagonal
graphene flakes $O(k, m, n)$ and oblate rectangles $Or(m, n)$, despite of substantial effort invested in
such a development [34,35,37,38,41,43–45]. We consider the problem of finding a closed formula
for $ZZ(O(k, m, n), x)$ with arbitrary values of the indices $k$, $m$, and $n$ the Holy Grail of the ZZ
polynomial theory. Other important aspects associated with the work on closed-form ZZ polynomial
formulas concern new theoretical concepts, ideas, and connections emerging during such work
and including among others a connection to transfer-matrix methodology [46], relations to cube
polynomials [47–49], a relation to tiling polynomials [50], the derivation of interface theory of
benzenoids [51–53], the development of the fragment and interface connectivity graphs [52–54],
a connection to John-Sachs theory of Kekulé structures [44,45], and a discovery of classes of benzenoids
for which the ZZ polynomials have only real zeros [55].

3. ZZ Polynomials of Complex Benzenoid Structures

We explained in the previous section that for most families of basic benzenoids, closed-form
expressions for their ZZ polynomials are known [18–20,24–29,33,36–40,53,54]. A topic that receives
more and more attention in the chemical graph theory community is the problem of how the Clar covers
of complex benzenoids built from basic benzenoids can be enumerated using as the counting blocks
the Holy Grail of the ZZ
polynomial theory. Other important aspects associated with the work on closed-form ZZ polynomial
formulas concern new theoretical concepts, ideas, and connections emerging during such work
and including among others a connection to transfer-matrix methodology [46], relations to cube
polynomials [47–49], a relation to tiling polynomials [50], the derivation of interface theory of
benzenoids [51–53], the development of the fragment and interface connectivity graphs [52–54],
a connection to John-Sachs theory of Kekulé structures [44,45], and a discovery of classes of benzenoids
for which the ZZ polynomials have only real zeros [55].
of ZZ polynomials of its simpler substructures [45], suggesting that it could be possible to formulate a
general John-Sachs theory of Clar covers.

In the current report, we want to analyze a slightly different problem. Instead of taking a complex
benzenoid and expressing it as a geometrical overlap of basic benzenoids, we rather select a collection
of simple, basic benzenoids and form all complex benzenoids that can be created by overlapping the
basic ones, and subsequently check if the ZZ polynomials of all of the resulting structures can be
expressed as simple functions of the ZZ polynomials of the basic structures. In particular, due to the
complexity of a general problem of this type, we analyze here all possible complex benzenoids that
can be formed by overlapping two identically oriented (but not identical) parallelograms, $M(m_1, n_1)$
and $M(m_2, n_2)$. A graphical list of all possible Kekuléan structures of this type is presented in Figure 3.
The next section reports closed-form formulas for the ZZ polynomials of all these structures together
with derivations and justifications of these formulas (whenever available). Four kinds of structures of
this type, presented in Figure 4, are obviously non-Kekuléan, as can be easily verified by checking that
their number of peaks $n_1$ and their number of valleys $n_\nu$ are not identical [16,57,58] (for more detailed
discussion, see for example p. 61 of [16]).
Figure 3. All types of Kekuléan composite benzenoids that can be formed by overlapping two identically oriented parallelogram-shaped benzenoids, \( M(m_1, n_1) \) and \( M(m_2, n_2) \). The parallelograms are depicted using blue and red shading and the overlapping region is depicted in purple.

Figure 4. All types of non-Kekuléan composite benzenoids that can be formed by overlapping two identically oriented parallelogram-shaped benzenoids, \( M(m_1, n_1) \) and \( M(m_2, n_2) \). The parallelograms are depicted using blue and red shading and the overlapping region is depicted in purple.

4. Results

4.1. Ribbons \( Rb (n_1, n_2 - n_1, m_2, m_1 - m_2) \), \( Rb (n_1, n_2 - n_1, m_2, m_1 - \mu) \), and \( Rb (n_1, n_2 - n_1, m_2, m_1) \)

The three structures \( Rb (n_1, n_2 - n_1, m_2, m_1 - m_2) \), \( Rb (n_1, n_2 - n_1, m_2, m_1 - \mu) \), and \( Rb (n_1, n_2 - n_1, m_2, m_1) \) shown in the bottom row of Figure 3 are usually referred to as generalized ribbons [58]. A closed-form formula of the ZZ polynomial of a ribbon was recently discovered [42] using recursive decompositions and interface theory considerations [51,52]. Adapting these results to the current case, we can give the following expression for the ZZ polynomials of the studied here generalized ribbons

\[
\text{ZZ}(Rb (n_1, n_2 - n_1, m_2, m_1 - \tau), x) = \sum_{k=0}^{\min(n_1, m_2)} \sum_{j=0}^{m_2-k} \sum_{i=0}^{n_1-k} (m_2-k)_j (n_2-n_1+k)_i (m_1-\tau+k)_i (n_1-1)_i (1 + x)^{i+j} \tag{4}
\]

\[
+ x \sum_{k=1}^{\min(n_1, m_2)} \sum_{j=0}^{m_2-k} \sum_{i=0}^{n_1-k} (m_2-k)_j (n_2-n_1-1+k)_i (m_1-\tau-1+k)_i (n_1-1)_i (1 + x)^{i+j}
\]
where the parameter $\tau$ assumes the following values: $\tau = m_2, \mu$, and 0 for the three structures shown from left to right, respectively, in the bottom row of Figure 3.

4.2. Two Vertically Overlapping Parallelograms $MvM(m_2, n_2, m_1, n_1, \mu, \nu)$

Let us denote by $MvM(m_2, n_2, m_1, n_1, \mu, \nu)$ the benzenoid obtained by overlapping vertically the parallelogram $M(m_2, n_2)$ located above the parallelogram $M(m_1, n_1)$ with the overlapping region $M(\mu, \nu)$. Before we proceed to the derivation, first we present basic tenets of this theory adapted here to $MvM$ oriented vertically.

Before we proceed to the determination of the ZZ polynomial of $MvM$, let us notice that three other structures of this type, $MvM(n_2, m_2, m_1, n_1, \mu, \nu)$, $MvM(m_1, m_1, m_2, n_2, \mu, \nu)$, and $MvM(n_1, m_1, n_2, m_2, \nu, \mu)$ all share the same ZZ polynomial as their sets of Clar covers can be obtained by an appropriate symmetry operation $\hat{S}$ (respectively: vertical reflection $\delta_{vert}$, horizontal reflection $\delta_{hor}$, or in-plane rotation $C_2$) from the set of Clar covers of the original structure. In the following, we consider these four structures as a single equivalence class and we choose a representative (briefly denoted as $MvM$) for this class such that $\mu \geq \nu$ and $m_1 \geq m_2$. Since the geometrical requirements for this class of structures dictate that $\mu < m_1, m_2$ and $\nu < n_1, n_2$, we always assume in the following that $m_1 \geq m_2 > \mu \geq \nu$ and $\nu < \min(n_1, n_2)$. The generalization of the obtained formulas to structures not satisfying the conditions $\mu \geq \nu$ and $m_1 \geq m_2$ will be given at the end of this Subsection.

The ZZ polynomial of $MvM$ can be derived from the interface theory of benzenoids [51,52]. Before we proceed to the derivation, first we present basic tenets of this theory adapted here to $MvM$ and necessary for completing this task.

- First we choose a standard orientation for $MvM$, analogous to the one shown in Figures 3 and 5 with the parallelogram $M(m_2, n_2)$ above $M(m_1, n_1)$ and a subset of edges of $MvM$ oriented vertically.
- We introduce in $MvM$ a system of parallel horizontal lines referred to as the elementary cuts $I_k$ in the way shown in Figure 5. Each such elementary cut is perpendicular to some vertical edges of $MvM$ and dissect them into halves. The number of elementary cuts introduced in this way is $n_2 + m_2 + n_1 + m_1 - \mu - \nu - 1$. For convenience, we augment this system with two additional elementary cuts, $I_0$ and $I_{n_2+m_2+n_1+m_1-\mu-\nu}$ in the way shown in Figure 5.
- The set of vertical edges of $MvM$ intersected by the elementary cut $I_k$ is referred to as the interface $i_k$. Each edge belonging to the interface $i_k$ is referred to as an interface bond. Simple geometrical considerations allow establishing that the number of interfaces in $MvM$ is $n_2 + m_2 + n_1 + m_1 - \mu - \nu - 1$. It is beneficial to augment this set again with two additional empty interfaces $i_0$ and $i_{n_2+m_2+n_1+m_1-\mu-\nu}$ in the way shown in Figure 5.
- The set of edges of $MvM$ located at least partially between the elementary cuts $I_k$ and $I_k$ is referred to as the fragment $f_k$. $MvM$ has $n_2 + m_2 + n_1 + m_1 - \mu - \nu$ fragments.
- For $k = 1, \ldots, n_2 + m_2 + n_1 + m_1 - \mu - \nu$, $i_{k-1}$ is the upper interface of $f_k$ and $i_k$ is the lower interface of $f_k$.
- Interface bonds in each fragment $f_k$ are numbered from left to right. The leftmost interface edge in $f_k$ is referred to as $e_{first} \equiv e_0$ and the rightmost interface edge in $f_k$, as $e_{last}$.
- Each fragment $f_k$ can be assigned an attribute of shape ($\equiv$ wider, $\equiv$ narrower, $R \equiv$ to-the-right, and $L \equiv$ to-the-left), which is defined in the following way

$$\text{shape}(f_k) = \begin{cases} 
\equiv & \text{when } e_{first} \in i_k \text{ and } e_{last} \in i_k \\
\equiv & \text{when } e_{first} \in i_{k-1} \text{ and } e_{last} \in i_k \\
\equiv & \text{when } e_{first} \in i_{k-1} \text{ and } e_{last} \in i_{k-1} \\
\equiv & \text{when } e_{first} \in i_k \text{ and } e_{last} \in i_{k-1} \\
\end{cases}$$

(5)

Following this convention, it is possible to assign the attribute of shape to the whole structure $MvM$, simply by listing the shape of each fragment from the top to the bottom. For example,
the shape sequences for the two structures \( MvM \) in Figure 5 are: (a) WWWRRNLWWWLLLNNNNNNN and (b) WWWRRBWWWRRINRRBRRNNNNNNN.

- Let us now consider an arbitrary Clar cover \( C \) of \( MvM \). For every interface edge \( e \) in \( MvM \), we define a covering order function \( \text{ord} \), which assumes the following values

\[
\text{ord} (e) = \begin{cases} 
1 & \text{when } \exists K_2 \in C : e \in E(K_2) \\
\frac{1}{2} & \text{when } \exists C_6 \in C : e \in E(C_6) \\
0 & \text{otherwise}
\end{cases}
\]  

(6)

where \( E(K_2) \) and \( E(C_6) \) denote the sets of edges of \( K_2 \) and \( C_6 \), respectively. We refer to a bond \( e \) with covering order 0 as a single bond, a bond \( e \) with covering order 1 as a double bond, and a bond \( e \) with covering order \( \frac{1}{2} \) as an aromatic bond.

- The concept of covering order (or briefly: order) can be naturally extended to interfaces. We define the covering order of the interface \( i \) as

\[
\text{ord} (i) = \sum_{e \in i} \text{ord} (e)
\]  

(7)

- Since the interface \( i_0 \) is empty, we naturally have \( \text{ord} (i_0) = 0 \). The orders of the remaining interfaces can be recursively computed from the First rule of interface theory \([51,52]\), which for an arbitrary Clar cover \( C \) relates the covering order of the interface \( i_k \) to the covering order of the interface \( i_{k-1} \) and the shape of the fragment \( f_k \) in the following way

(a) If \( f_k \) has the shape \( \mathbb{W} \), then \( \text{ord} (i_k) = \text{ord} (i_{k-1}) + 1 \).

(b) If \( f_k \) has the shape \( \mathbb{L} \), then \( \text{ord} (i_k) = \text{ord} (i_{k-1}) - 1 \).

(c) If \( f_k \) has the shape \( \mathbb{R} \) or \( \mathbb{L} \), then \( \text{ord} (i_k) = \text{ord} (i_{k-1}) \).

- The interface orders obtained in this way are actually independent of the choice of the Clar cover \( C \), as they are completely determined by the condition \( \text{ord} (i_0) = 0 \) and the shape sequence WWWRRNLWWWLLLNNNNNNN. Therefore, the interface orders are identical for every Clar cover \( C \) of \( MvM \) and can be treated as an inherent property of \( MvM \) allowing enumerating and constructing the set of Clar covers of \( MvM \). The interface covering orders computed in this way are listed in red for the two structures \( MvM \) shown in Figure 5.

- The number of interface bonds in every non-empty interface \( i_k \) of \( MvM \) is larger by 1 from the order of this interface, \( \text{ord} (i_k) \), as can be easily seen from Figure 5. This property holds for a general structure of this type, as both the interface orders and the numbers of interface bonds in consecutive interfaces depend in the same manner on the shape of the fragment between the interfaces, except for the first and the last fragment.

- An explicit formula for the interface order \( \text{ord} (i_k) \) as a function of the interface number \( k \) is somewhat cumbersome. It can be shown that

\[
\text{ord} (i_k) = p_1 (k) + p_2 (k) - p_{12} (k),
\]  

(8)

where

\[
p_1 (k) = \begin{cases} 
0 & \text{when } k < m_2 + n_2 + 1 - \mu - \nu \\
k - m_2 - n_2 + \mu + \nu & \text{when } m_2 + n_2 + 1 - \mu - \nu \leq k < m_2 + n_2 - \mu - \nu + \min(m_1, n_1) \\
\min(m_1, n_1) & \text{when } m_2 + n_2 - \mu - \nu + \min(m_1, n_1) \leq k \leq m_2 + n_2 - \mu - \nu + \max(m_1, n_1) \\
m_2 + n_2 + m_1 + n_1 - \mu - \nu - k & \text{when } k > m_2 + n_2 - \mu - \nu + \max(m_1, n_1)
\end{cases}
\]  

(9)
with an opportunity for designing an algorithm to determine a closed-form ZZ polynomial formula. The interface bonds are determined from Equation (8). For the two structures \( M(m_1, n_1) \), \( M(m_2, n_2) \), and \( M(\mu, \nu) \), respectively.

\[
p_2(k) = \begin{cases} 
  k & \text{when } k < \min(m_2, n_2) \\
  \min(m_2, n_2) & \text{when } \min(m_2, n_2) \leq k < \max(m_2, n_2) \\
  m_2 + n_2 - k & \text{when } \max(m_2, n_2) \leq k \leq m_2 + n_2 - 1 \\
  0 & \text{when } k > m_2 + n_2 - 1
\end{cases}
\]  

\[
p_{12}(k) = \begin{cases} 
  0 & \text{when } k < m_2 + n_2 + 1 - \mu - \nu \\
  k - m_2 - n_2 + \mu + \nu & \text{when } m_2 + n_2 + 1 - \mu - \nu \leq k < m_2 + n_2 + \mu - \nu \\
  \min(\mu, \nu) & \text{when } m_2 + n_2 + \mu - \nu \leq k < m_2 + n_2 + \nu - \mu \\
  m_2 + n_2 - k & \text{when } m_2 + n_2 - \nu \leq k \leq m_2 + n_2 - 1 \\
  0 & \text{when } k > m_2 + n_2 - 1
\end{cases}
\]  

where \( p_1(k) \), \( p_2(k) \), and \( p_{12}(k) \) correspond to the order of the same interface in the pristine structures \( M(m_1, n_1) \), \( M(m_2, n_2) \), and \( M(\mu, \nu) \), respectively.

![Figure 5](image-url)

**Figure 5.** Elementary cuts (in gray), interface orders (in red), and the numbers of interface bonds (in blue) for each interface \( i_k \) of (a) two vertically overlapping parallelograms \( M(7, 5) \) and \( M(7, 10) \) with the intersection region corresponding to the parallelogram \( M(4, 3) \) and (b) two vertically overlapping parallelograms \( M(12, 4) \) and \( M(13, 5) \) with the intersection region corresponding to the parallelogram \( M(9, 2) \). The interface bonds are defined as the bonds intersected by the elementary cuts. The interface \( i_k \) is defined as the set of interface bonds intersected by the same elementary cut \( i_k \). The depicted-in-blue interface cuts \( I_6 \) and \( I_9 \) (left panel) and \( I_7 \) and \( I_{14} \) (right panel) are used to partition the studied structures into smaller substructures, which allows us to derive closed-form formulas of their ZZ polynomials. For details, see text.

The deduced so far geometric properties and interface covering characters of \( M \vee M \) present us with an opportunity for designing an algorithm to determine a closed-form ZZ polynomial formula for this structure. To this end, let us consider explicitly the interface coverings of two particular interfaces of \( M \vee M \), \( I_{n_2 + m_2 - \mu} \) and \( I_{n_2 + m_2 - \nu} \). These two interfaces—intersected by the blue elementary cuts \( I_6 \) and \( I_9 \) (a) and \( I_7 \) and \( I_{14} \) (b) in Figure 5—can be characterized as the interfaces containing, respectively, the leftmost and rightmost interface bonds of the intersection region \( M(\mu, \nu) \). Let us denote by \( \beta_\mu \equiv \text{ord}(I_{n_2 + m_2 - \mu}) \) and \( \beta_\nu \equiv \text{ord}(I_{n_2 + m_2 - \nu}) \) the interface orders of these two interfaces as determined from Equation (8). For the two structures \( M \vee M \) shown in Figure 5, the interface orders computed in this way are \( \beta_\mu = 4 \) and \( \beta_\nu = 4 \) (a) and \( \beta_\mu = 4 \) and \( \beta_\nu = 5 \) (b). For a general structure \( M \vee M \), it can be shown that Equation (8) reduces to

\[
\beta_\mu = \min(\mu, n_2),
\]  

\[
\beta_\nu = \min(\mu, n_1).
\]
Let us now construct all possible interface coverings for the interface $i_{n_2+m_2-\mu}$. Following the discussion above, we can construct $o_\mu + 1$ distinct coverings of the interface $i_{n_2+m_2-\mu}$ corresponding to the partition $1 + \ldots + 1 + 0$ and $o_\mu$ distinct coverings corresponding to the partition $1 + \ldots + 1 + \frac{1}{2} + \frac{1}{2}$. In the first case, the multiset of interface bonds covering orders consists of $o_\mu$ 1s and one 0; the distinction between Clar covers belonging to this category concerns the location of the single bond. Similarly, in the second case, the multiset of interface bonds covering orders consists of $o_\mu - 1$ 1s and two $\frac{1}{2}$s with the distinction between Clar covers belonging to this category concerning the location of the two consecutive aromatic bonds. Each of these $2 o_\mu + 1$ partitions generates a system of fixed bonds in $MvM$ not limited only to interface $i_{n_2+m_2-\nu}$ but extending through both overlapping parallelograms. The systems of fixed bonds associated with each of these partitions for the structure (a) from Figure 5 is shown in Figure 6. This system of fixed bonds is a simple consequence of the Second rule of interface theory [51,52], which—figuratively speaking—ensures that consecutive covered interface bonds in each fragment $f_k$ alternate between the upper and lower interfaces, $i_{k-1}$ and $i_k$. The system of fixed bonds propagates to upper and lower interfaces of $MvM$ in the following way forming two disconnected, wedge-shaped, non-fixed subgraphs of $MvM$, one located above the interface $i_{n_2+m_2-\nu}$ and one below.

In case when $i_{n_2+m_2-\mu} = i_{n_2+m_2-\nu}$ (i.e., when $\mu = \nu$), the two disconnected, wedge-shaped, non-fixed subgraphs of $MvM$ formed by the system of fixed bonds both have the shape of a parallelogram. In addition, the order of the interface $i_{n_2+m_2-\mu}$ reduces to $o_\mu = \mu$. It is easy to deduce that by selecting the partition $1 + \ldots + 1 + 0$ with the single bond located in position $k$ ($k = 0, \ldots, \mu$), the lower parallelogram corresponds to $M(m_1 - k, n_1 - \nu + k)$ and the upper to $M(m_2 - \mu + k, n_2 - k)$. For example, for the structure $MvM$ obtained by merging vertically $M(6,8)$ with $M(6,6)$ by $M(3,3)$, we obtain four pairs of such parallelograms.

Similarly, for the partition $1 + \ldots + 1 + \frac{1}{2} + \frac{1}{2}$ with an aromatic ring in hexagon $k$ ($k = 1, \ldots, \mu$), the lower parallelogram corresponds to $M(m_1 - k, n_1 - \nu + k - 1)$ and the upper to $M(m_2 - \mu + k - 1, n_2 - k)$. For the same structure $MvM$ analyzed a few lines above we obtain three pairs of such parallelograms.
Consequently, the total set of Clar cover of $MvM$ can be partitioned into $2 \mu + 1$ disjoint sets, each of them corresponding to one of the $1 + \ldots + 1 + 0$ or $1 + \ldots + 1 + \frac{1}{2} + \frac{1}{2}$ partitions. The ZZ polynomial of $MvM$ can be then constructed as a sum of ZZ polynomials of the non-fixed disconnected components of $MvM$ over all the partition classes

$$ZZ(MvM, x) = \sum_{k=0}^{\mu} ZZ(M(m_2 - \mu + k, n_2 - k), x) \cdot ZZ(M(m_1 - k, n_1 - \nu + k), x)$$  \hspace{1cm} (14)

$$+ \sum_{k=1}^{\mu} x \cdot ZZ(M(m_2 - \mu + k - 1, n_2 - k), x) \cdot ZZ(M(m_1 - k, n_1 - \nu + k - 1), x)$$

where the additional factor $x$ in the second line of Equation (14) symbolizes the aromatic ring in the fixed component of $MvM$ for each $1 + \ldots + 1 + \frac{1}{2} + \frac{1}{2}$ partition class. Since both lines of this formula are isostructural, we are able to write Equation (14) in even more compact form as

$$ZZ(MvM, x) = \sum_{i=0}^{1} \sum_{k=0}^{\mu} ZZ(M(m_2 - \mu + k + i, n_2 - k), x) \cdot ZZ(M(m_1 - k, n_1 - \nu + k + i), x) \cdot x^i$$  \hspace{1cm} (15)

**Figure 6.** Assigning bond coverings to the interface $i_8$ of the structure (a) from Figure 5 results in nine different systems of fixed bonds (depicted in gray). Five of these systems correspond to four double bonds and one single bond (upper row, partitions $1 + \ldots + 1 + 0$) and four of these systems correspond to one aromatic ring and three double bonds (lower row, partitions $1 + \ldots + 1 + \frac{1}{2} + \frac{1}{2}$). The non-fixed components of each graph (depicted in black) correspond to two disconnected subgraphs of $MvM$, here both with the shape of a parallelogram. The shapes of the parallelograms are determined by the position of the single bond or by the position of the aromatic ring.
In the case when \( i_{n_2+m_2-\mu} \neq i_{n_2+m_2-\nu} \) (i.e., when \( \mu > \nu \)), to obtain a formula for the ZZ polynomial of \( M\nu M \) analogous to Equation (14), we need to consider all the combinations of the coverings of the interfaces \( i_{n_2+m_2-\mu} \) and \( i_{n_2+m_2-\nu} \) simultaneously. Two important aspects of this process need to be addressed: (1) Not every covering of \( i_{n_2+m_2-\mu} \) is compatible with every covering of \( i_{n_2+m_2-\nu} \) (and vice versa); the precise compatibility conditions and their origin are discussed below. (2) A selection of compatible coverings of both interfaces induces a system of fixed bonds in \( M\nu M \), which separates three disconnected, parallelogram-shaped, non-fixed subgraphs of \( M\nu M \). The actual dimensions of the parallelograms, depending on the positions of the single bond(s) and/or aromatic ring(s) in both interface coverings, are derived below. As an illustration we show in Figure 7 four different systems of fixed bonds (depicted in gray) and non-fixed bonds (depicted in black) associated with four selected covering types of the interfaces \( i_8 \) and \( i_{14} \) in \( M\nu M \) formed by overlapping vertically \( M(11,6) \) with \( M(11,9) \) by \( M(9,3) \). Note that in the leftmost covering, the central parallelogram located between both interfaces formally corresponds to \( M(0,5) \) and allows only a single Clar cover; therefore it can be treated either as a fixed-bond region or non-fixed-bond region.

![Figure 7](image)

**Figure 7.** Four examples of partially covered structures \( M\nu M(11,6,11,9,9,3) \) show that the non-covered regions consists of three parallelograms with their shapes determined by the location of the single and/or aromatic bonds in the both covered interfaces \( i_{n_2+m_2-\mu} \) and \( i_{n_2+m_2-\nu} \).

In this paragraph we discuss the compatibility conditions of both interface coverings. As already mentioned above, selecting a particular covering of \( i_{n_2+m_2-\mu} \) induces a system of fixed bonds in \( M\nu M \), which extends vertically to the other interface, \( i_{n_2+m_2-\nu} \), imposing double bond covering of some of its bonds. The available range of non-fixed interface bonds in \( i_{n_2+m_2-\nu} \) can be characterized as follows.

- Let us assume that a \( 1 + \ldots + 1 + 0 \) partition was selected for \( i_{n_2+m_2-\mu} \) with the single bond in position \( k \in [0, \ldots, o_\mu] \). Somewhat involved geometric considerations show that the range of indices of non-fixed interface bonds in \( i_{n_2+m_2-\nu} \) associated with this choice is given by \([\max(k + o_\nu - \mu, 0), o_\nu - \max(0, v - k)]\).
- Consequently, a single bond in interface \( i_{n_2+m_2-\mu} \) in position \( k \) permits placing a single bond in interface \( i_{n_2+m_2-\nu} \) in position \( l \in [\max(k + o_\nu - \mu, 0), o_\nu - \max(0, v - k)] \) or permits placing an aromatic ring in hexagon \( l \in [1 + \max(k + o_\nu - \mu, 0), o_\nu - \max(0, v - k)] \).
- Let us assume now that a \( 1 + \ldots + 1 + \frac{1}{2} + \frac{1}{2} \) partition was selected for \( i_{n_2+m_2-\mu} \) with the aromatic ring in hexagon \( k \in [1, \ldots, o_\mu] \). Again, geometric considerations show that the range of indices of non-fixed interface bonds in \( i_{n_2+m_2-\nu} \) associated with this choice of covering for \( i_{n_2+m_2-\mu} \) is given by \([\max(k + o_\nu - \mu, 0), o_\nu - \max(0, v - k + 1)]\).
- Consequently, an aromatic ring in hexagon \( k \) of interface \( i_{n_2+m_2-\mu} \) permits placing a single bond in interface \( i_{n_2+m_2-\nu} \) in position \( l \in [\max(k + o_\nu - \mu, 0), o_\nu - \max(0, v - k + 1)] \) or permits placing an aromatic ring in hexagon \( l \in [1 + \max(k + o_\nu - \mu, 0), o_\nu - \max(0, v - k + 1)] \).

Let us now give the dimensions for the three parallelograms induced by the systems of fixed bonds in \( M\nu M \) associated with placing a single bond \( (i = 0) \) or an aromatic ring \( (i = 1) \) in position \( k \) in the interface \( i_{n_2+m_2-\mu} \) and a single bond \( (j = 0) \) or an aromatic ring \( (j = 1) \) in position \( l \) in the interface \( i_{n_2+m_2-\nu} \). It is possible to demonstrate via elementary geometric considerations that in this
situation the upper parallelogram $M_u \equiv M_{upper}$, the lower parallelogram $M_l \equiv M_{lower}$, and the middle parallelogram $M_m \equiv M_{middle}$ are given by the following expressions

$$
M_u = M(m_2 - \mu + k - j, n_2 - k) \tag{16}
$$

$$
M_l = M(m_1 + o_\nu - \mu - l, n_1 - o_\nu + l - i) \tag{17}
$$

$$
M_m = M(\mu - o_\nu + l - k - i, o_\nu - \nu - l + k - j) \tag{18}
$$

Consequently, the ZZ polynomial of $\mathbb{M}M$ with $\mu > \nu$ is given by the following expression

$$
ZZ(\mathbb{M}M, x) = \sum_{i=0}^{1} \sum_{j=0}^{1} \sum_{k=0}^{\nu - \mu - 0} \sum_{l=0}^{\nu - \mu - 0} a(\tau) \cdot z(\tau) \cdot x^{i+j} (19)
$$

where explicit expressions for $ZZ(M_u, x)$, $ZZ(M_m, x)$, and $ZZ(M_l, x)$ are given by Equation (2). Substituting these expressions into Equation (19) produces a formula with a septuple summation and six binomial coefficients, which is not given explicitly here.

As mentioned earlier, the structure $\mathbb{M}M$ with $\mu \geq \nu$ and $m_1 \geq m_2$, for which the ZZ polynomial formula is given by Equation (19) (when $\mu > \nu$) and by Equation (15) (when $\mu = \nu$), is a representative for an equivalence class of four structures sharing the same ZZ polynomial. We have thus two possible pathways leading to the determination of the ZZ polynomial for a general structure $\mathbb{M}M\mathbb{M}(m_2, n_2, m_1, n_1, \mu, \nu)$, for which the conditions $\mu \geq \nu$ and $m_1 \geq m_2$ are not satisfied. The first natural way is to identify the equivalence class representative $\mathbb{M}M$ for $\mathbb{M}M\mathbb{M}(m_2, n_2, m_1, n_1, \mu, \nu)$ by producing four symmetry-related structures ($\bar{E}(\mathbb{M}M\mathbb{M}(m_2, n_2, m_1, n_1, \mu, \nu))$, $\bar{C}_2(\mathbb{M}M\mathbb{M}(m_2, n_2, m_1, n_1, \mu, \nu))$, $\bar{\delta}_{\text{box}}(\mathbb{M}M\mathbb{M}(m_2, n_2, m_1, n_1, \mu, \nu))$, and $\bar{\delta}_{\text{vert}}(\mathbb{M}M\mathbb{M}(m_2, n_2, m_1, n_1, \mu, \nu))$, where $D_2 = \{\bar{E}, \bar{C}_2, \bar{\delta}_{\text{box}}, \bar{\delta}_{\text{vert}}\}$ is the 2D dihedral group) and selecting a structure for which $\mu \geq \nu$ and $m_1 \geq m_2$. Perhaps such a solution is aesthetically pleasing, but from a practical standpoint it is more convenient to use a symmetry-adapted version of Equations (15) and (19) given by the following expression

$$
ZZ(\mathbb{M}M, x) = \begin{cases} 
\sum_{i=0}^{1} \sum_{k=0}^{\nu - \mu - 0} ZZ(M_u, x) \cdot ZZ(M_b, x) \cdot x^{i} & \text{when } \mu = \nu \\
\sum_{i=0}^{1} \sum_{j=0}^{1} \sum_{k=0}^{\nu - \mu - 0} ZZ(M_u, x) \cdot ZZ(M_m, x) \cdot ZZ(M_l, x) \cdot x^{i+j} & \text{when } \mu \neq \nu
\end{cases} \tag{20}
$$

where $\mathbb{M}M$ stands already for an arbitrary structure $\mathbb{M}M\mathbb{M}(m_2, n_2, m_1, n_1, \mu, \nu)$ and where the following symbols were used

$$
o_{\mu} = \begin{cases} 
\min(\mu, n_2) & \text{when } \mu > \nu \\
\min(\nu, m_2) & \text{when } \nu > \mu
\end{cases} \tag{21}
$$

$$
o_{\nu} = \begin{cases} 
\min(\mu, n_1) & \text{when } \mu > \nu \\
\min(\nu, m_1) & \text{when } \nu > \mu
\end{cases} \tag{22}
$$

$$
a(\tau) = \begin{cases} 
\max(\tau + o_{\nu} - \mu, 0) & \text{when } \mu > \nu \\
\max(\tau + o_{\nu} - \nu, 0) & \text{when } \nu > \mu
\end{cases} \tag{23}
$$

$$
z(\tau) = \begin{cases} 
o_{\nu} - \max(0, \nu - \tau) & \text{when } \mu > \nu \\
o_{\nu} - \max(0, \mu - \tau) & \text{when } \nu > \mu
\end{cases} \tag{24}
$$
whose diagonal elements

while the MAPLE procedure produces identical results instantaneously.

provided MAPLE procedure. Note that the computational time needed to evaluate the ZZ polynomials
of larger structures using recursive decompositions with ZZDecomposer exceeds a few minutes,

MvM

in Figure 8 a MAPLE procedure

MhM

A closed-form formula for the ZZ polynomial of the composite benzenoid

\( M_{\text{hM}}(m_1, n_1, m_2, n_2, \mu, \nu) \)

shown schematically in Figure 3 has not been reported yet. Extensive
numerical experimentation shows that the appropriate formula can be written as a determinant of a
certain 2 \times 2 matrix

\[
\text{ZZ}(M_{\text{hM}}(m_1, n_1, m_2, n_2, \mu, \nu), x) = \begin{vmatrix} t_1 & u \\ v & t_2 \end{vmatrix}
\]

(30)

whose diagonal elements \( t_1 \) and \( t_2 \) correspond to the ZZ polynomials of the parallelograms \( M(m_1, n_1) \)
and \( M(m_2, n_2) \), respectively,

\[
t_1 = \text{ZZ}(M(m_1, n_1), x) = \sum_{k=0}^{\min(m_1, n_1)} (\binom{m_1}{k})(\binom{n_1}{k})(1+x)^k
\]

(31)

\[
t_2 = \text{ZZ}(M(m_2, n_2), x) = \sum_{k=0}^{\min(m_2, n_2)} (\binom{m_2}{k})(\binom{n_2}{k})(1+x)^k
\]

(32)

and the off-diagonal elements \( u \) and \( v \) are given explicitly by the following expressions

\[
u = \sum_{k=0}^{\mu} (\binom{m_1+n_2-k-2}{k+2})(1+x)^{k+1}
\]

(33)

\[
u = \sum_{k=0}^{\mu} (\binom{n_1+m_2-v-2}{k+2})(1+x)^{k+1}
\]

(34)

We have no formal proof demonstrating the correctness of Equation (30), but at the same time
the overwhelming numerical evidence clearly shows that Equation (30) is correct and applicable to
structures \( M_{\text{hM}}(m_1, n_1, m_2, n_2, \mu, \nu) \) with arbitrary values \( m_1, m_2 > \mu \geq 0 \) and \( n_1, n_2 > \nu \geq 0 \) of the
structural parameters.
Figure 8. Maple procedure for computing the ZZ polynomial of an arbitrary structure $MvM \equiv MvM(m_2, n_2, m_1, n_1, \mu, \nu)$ according to Equations (20)–(29). As a numerical verification of correctness of the derived formula, several typical structures $MvM$ is shown together with their ZZ polynomials evaluated in a twofold manner: with ZZDecomposer (in black) and with the provided MAPLE routine (in blue in red frames).

It might be instructive to describe briefly the thought process leading to the discovery of Equation (30). In 1952 Gordon and Davison observed that there is a one-to-one correspondence between the number of Kekulé structures for a given benzenoid $B$ and the number of monotonic path systems in $B$ [59]. This results was rigorously proved in 1984 by Sachs [60]. Subsequently, John, Sachs, and Rempel [61,62] and later Cyvin and Gutman [57,58,63] designed a simple strategy allowing computing the Kekulé count of $B$ by taking a determinant of the so-called John-Sachs path matrix. The John-Sachs path matrix is a square matrix of size $p \times p$, where $p$ is the number of peaks (or equivalently, valleys) in the Kekuléan benzenoid $B$. The rows of the matrix are associated with the peaks of $B$ and the columns, with the valleys of $B$. The $(i, j)$ entry of the matrix is the Kekulé count for the subgraph of $B$ containing only those vertices of $B$ that can be reached from the peak $p_i$ by going exclusively downward and simultaneously that can be reached from the valley $v_j$ by going exclusively upward. For details, we refer the reader to the excellent exposition with multiple examples given by Gutman and Cyvin [63] or by Bodroža and collaborators [43]. For the
studied here composite benzenoid \( M_{hh}(m_1, n_1, m_2, n_2, \mu, v) \), the John-Sachs path matrix is of size \( 2 \times 2 \), because this structure contains 2 peaks and 2 valleys. The diagonal elements of this matrix correspond to the Kekulé counts of \( M (m_1, n_1) \) and \( M (m_2, n_2) \), and the off-diagonal elements, to the Kekulé counts of \( M (m_1 + m_2 - \mu, v) \) and \( M (\mu, n_1 + n_2 - v) \). We asked ourselves whether it is possible to extend the John-Sachs path matrix construction to the problem of determination of ZZ polynomial for this structure and to our surprise we discovered that indeed such an extension can be constructed. The diagonal entries of the original John-Sachs matrix are naturally replaced by the ZZ polynomials of the \((p_1, v_1)\) subgraphs (i.e., by \( ZZ (M (m_1, n_1), x) \) and \( ZZ (M (m_2, n_2), x) \)), which are given explicitly by \( t_1 \) and \( t_2 \) in Equations (31) and (32). Discovering the actual analytic form of the off-diagonal elements \( u \) and \( v \) requires quite an extensive experimentation, but the expressions given by Equations (33) and (34) are not surprising and show a clear connection to the \( (p_i, v_j) \) subgraphs (i.e., to \( M (m_1 + m_2 - \mu, v) \) and \( M (\mu, n_1 + n_2 - v) \), even if the details of this connection are quite murky at the time and the presence of the multiplicative factor \((1 + x)\) and the positive and negative shift of 2 in the parallelograms dimensions escapes our understanding. The presented here example of extending the John-Sachs theory of Kekulé structures to the world of Clar covers suggests that there might exist a generalization of the John-Sachs theorem valid not only for monotonic paths consisting of the \( K_2 \) graphs, but also for generalized monotonic paths consisting of the \( K_2 \) and \( C_6 \) graphs. Note that once such a general theory of general monotonic path systems is conceived, it will provide a proper justification to the conjecture expressed by Equation (30).

### 4.4. Two Intersecting Parallelograms \( M_{xx}(m_2, n_2, m_1, n_1, \mu, v) \)

A union \( M_{xx}(m_2, n_2, m_1, n_1, \mu, v) \) of two intersecting parallelograms \( M (m_1, n_1) \) and \( M (m_2, n_2) \), shown schematically in Figure 9, can be formally treated as a union of two overlapping ribbons, \( Rb (n_1, v, m_2, \mu) \) and \( Rb (n_1, n_2 - n_1 - v, m_2, m_1 - m_2 - \mu) \), where the first ribbon was rotated by \( \pi \) with respect to the standard orientation [42,58]. This second interpretation allows us to use exactly the same technique as in the last subsection to find the ZZ polynomial formula for \( M_{xx}(m_2, n_2, m_1, n_1, \mu, v) \).

We again seek for an appropriate ZZ polynomial formula in the form of \( 2 \times 2 \) determinant, with the diagonal elements \( t_1 \) and \( t_2 \) corresponding to the ZZ polynomials of ribbons \( Rb (n_1, v, m_2, \mu) \) and \( Rb (n_1, n_2 - n_1 - v, m_2, m_1 - m_2 - \mu) \) and the off-diagonal elements \( u \) and \( v \) somehow related to the two intersecting parallelograms \( M (m_1, n_1) \) and \( M (m_2, n_2) \). Numerical experimentation shows that indeed the sought ZZ polynomial can be represented in the anticipated way as

\[
ZZ(M_{xx}(m_2, n_2, m_1, n_1, \mu, v), x) = \begin{vmatrix} t_1 & u \\ v & t_2 \end{vmatrix}
\]

with \( u, v, t_1, \) and \( t_2 \) given explicitly by the following expressions

\[
u = \sum_{k=0}^{v} \binom{m_1 - 2}{k} \binom{n_1 + 2}{k+2} (1 + x)^{(k+1)}
\]

\[
v = \sum_{k=0}^{\mu} \binom{m_2 - 2}{k} \binom{n_2 + 2}{k+2} (1 + x)^{(k+1)}
\]

\[
t_1 = ZZ (Rb (n_1, v, m_2, \mu), x)
\]

\[
= \sum_{k=0}^{\min(m_1, m_2)} \sum_{j=0}^{m_2 - k} \sum_{i=0}^{n_1 - k} \binom{m_2 - k}{j} \binom{n_1 - k}{i} (1 + x)^{i+j}
\]

\[
+ x \sum_{k=1}^{\min(m_1, m_2)} \sum_{j=0}^{m_2 - k} \sum_{i=0}^{n_1 - k} \binom{m_2 - k}{j} \binom{n_1 - k}{i} (1 + x)^{i+j}
\]
\[ t_2 = \text{ZZ} \left( Rb \left( n_1, n_2 - n_1 - \nu, m_2, m_1 - m_2 - \mu \right), x \right) \]
\[ = \sum_{k=0}^{\min(n_1,m_2)} \sum_{j=0}^{n_2-k} \sum_{i=0}^{n_1-k} (m_2-k) (n_2-n_1-\nu+k) (m_1-m_2-\mu+k) (n_1-k)(1+x)^{i+j} \]
\[ + x \sum_{k=1}^{\min(n_1,m_2)} \sum_{j=0}^{n_2-k} \sum_{i=0}^{n_1-k} (m_2-k) (n_2-n_1-\nu-1+k) (m_1-m_2-\mu-1+k) (n_1-k)(1+x)^{i+j} \]

Again, we have no formal proof demonstrating the correctness of Equation (35) but at the same time multiple numerical checks convince us that Equation (35) is correct and applicable to all the composite benzenoids \( MxM(m_2, n_2, m_1, n_1, \mu, \nu) \) with the structural parameters satisfying the following conditions: \( \mu, \nu > 0, n_2 > n_1 + \nu, \) and \( m_1 > m_2 + \mu. \) Following the discussion at the end of Section 4.3, we believe that Equation (35) will remain a conjecture until a theory of general monotonic path systems (i.e., an extension of John-Sachs theory applicable to Clar covers) is developed.

Figure 9. The composite benzenoid \( MxM(m_2, n_2, m_1, n_1, \mu, \nu) \) shown originally in Figure 3 can be formally treated as a superposition of two parallelograms, \( M(m_1, n_1) \) (red shading) and \( M(m_2, n_2) \) (blue shading), or two ribbons, \( Rb(n_1, \nu, m_2, \mu) \) (red shading) and \( Rb(n_1, n_2 - n_1 - \nu, m_2, m_1 - m_2 - \mu) \) (blue shading).

5. Conclusions

We present a complete set of closed-form formulas for the ZZ polynomials of composite benzenoid structures obtained by overlapping two parallelograms, \( M(m_1, n_1) \) and \( M(m_2, n_2) \). Depending on the mutual location of the parallelograms and the dimension of the overlap parallelogram-shaped region, \( M(\mu, \nu) \), the resulting composite structure can be Kekuléan (see Figure 3) or non-Kekuléan (see Figure 4). We identify five families of Kekuléan structures: parallelograms \( M(m_1 + m_2 - \mu, n_1 = n_2) \) and \( M(m_1 = m_2, n_1 + n_2 - \nu) \) with ZZ polynomials given by Equation (2), generalized ribbons \( Rb \) with ZZ polynomials given by Equation (4), horizontally overlapping parallelograms \( MhM(m_1, n_1, m_2, n_2, \mu, \nu) \) with ZZ polynomials given by Equation (30), intersecting parallelograms \( MxM(m_2, n_2, m_1, n_1, \mu, \nu) \) with ZZ polynomials given by Equation (35), and vertically overlapping parallelograms \( MvM(m_2, n_2, m_1, n_1, \mu, \nu) \) with ZZ polynomials given by Equation (20). All formulas are given in the form of multiple sums over binomial coefficients, bearing a close structural similarity to the ZZ polynomial of a pristine parallelogram \( M(m, n) \) given by Equation (2). In case of the subset of the composite benzenoids consisting of parallelograms \( M, \) generalized ribbons \( Rb, \) and vertically overlapping parallelograms \( MvM, \) we demonstrate formally the correctness of the derived formulas. For intersecting parallelograms \( MxM \) and horizontally overlapping parallelograms \( MhM \) the derived formulas are given without a proof, in the form of conjectures; pervasive numerical evidence suggests correctness of these conjectures. Since both the conjectured formulas have the form of a \( 2 \times 2 \)
determinant with a striking resemblance to the John-Sachs determinantal formulas for the number of Kekulé structures [57,58,61–63], we expect that their formal proof could be possible only once the John-Sachs theory of Kekulé structures is generalized to the world of Clar covers. We hope that our results will stimulate the graph theoretical community to accomplish this task and simultaneously will provide an appropriate testing ground for such a generalization. The formal demonstration of correctness of the ZZ polynomial formulas for generalized ribbons and vertically overlapping parallelograms is based on the recently announced interface theory of benzenoids [51–53], which seems to be a promising tool for solving numerous challenging problems in chemical graph theory. All the derived and reported here ZZ polynomial formulas were tested against numerous instances of actual composite structures of a given type, for which the ZZ polynomials were computed in a brute-force decomposition manner using ZZDecomposer [25–27].

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