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ORIGINAL PAPER

Topological chirality of a type of DNA and protein polyhedral links

Xian'an Jin[1] · Xiao-Sheng Cheng[1,2]

Abstract Polyhedral links, interlinked and interlocked architectures, have been proposed for the description and characterization of DNA and protein polyhedra. In this paper, we study the topological chirality of a type of DNA polyhedral links constructed by the strategy of "n-point stars" and a type of protein polyhedral links constructed by "three-cross curves and untwisted double-line" covering. Furthermore, we prove that links corresponding to bipartite plane graphs have antiparallel orientations, and under these orientations, their writhes are not zero. As a result, the type of double crossover DNA polyhedral links are topologically chiral. We also prove that the unoriented link corresponding to a connected, even, bipartite plane graph always has self-writhe 0. Using the Jones polynomial for unoriented links we derive two simple criteria for chirality of unoriented alternating links with self-writhe 0. By applying this criterion we show that 3-regular protein polyhedral links are also topologically chiral. Topological chirality always implies chemical chirality, hence the corresponding DNA and protein polyhedra are all chemically chiral. Our chiral criteria may be used to detect the topological chirality of more complicated DNA and protein polyhedral links that may be synthesized by chemists and biologists in the future.

Keywords Chirality criteria · Polyhedral links · DNA and protein backbone · Plane graphs

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1 Introduction

Chirality is an important feature of all biomacromolecules, which are responsible for the structure and chemical regulation of living cells and DNA. Most biomolecules, such as amino acids, sugars, RNA, DNA and proteins are chiral. Knotted DNA and proteins are intriguing. Scientists have found and synthesized many DNA and protein knots and links. For example, DNA trefoil [1], DNA Whitehead link and 6_2 knot [2], and many more examples of DNA knots can be found in [3,4]; protein figure-eight knot [5], protein 5_2 knot [6], and in fact, 3–6 crossing twist knots have been found in protein. For a list of such links, see [7].

In the last 20 years or so, many DNA and protein biomolecules with the shape of polyhedron have been synthesized and found. For example, DNA cube [8], DNA tetrahedron [9], DNA octahedron [10], DNA truncated octahedron [11], DNA bipyramid [12], DNA dodecahedron [13]. They are all constructed by means of "n-branched curves and even-twisted double-line" covering and the chirality of their corresponding polyhedral links was studied in [14]. In recent several years, a type of more complicated DNA polyhedra have been reported in [15–18]. They are all constructed by the strategy of "n-point stars" and called double crossover DNA polyhedra in [19]. Similar DNA molecular structures can be found in [20,21]. In [15], the chirality of double crossover DNA octahedron was considered, but in general the chirality of this type of polyhedra has not been systematically analyzed. Proteins as building blocks were also used to form polyhedral structures. As far as we know, only one protein polyhedral structure has been found in the laboratory, that is the HK97 capsid [22]. Such polyhedra can be modelled mathematically by 3-regular polyhedral links via "three-cross curves and untwisted double-line" covering [23,24]. The chirality of 3-regular polyhedral links was studied in [25].

We recall that a molecule is chemically chiral if its structure cannot be deformed into its mirror-image structure by realizable intramolecular motions. If we view a molecular structure as a rigid object and can deform it to its mirror image by moving it in the three-dimensional space, we call it geometrically achiral and geometrically chiral otherwise. If we view a molecular structure as a completely elastic object and can deform it to its mirror image, we call it topologically achiral and topologically chiral otherwise. The relation among these three chiralities is summarized as follows: topological chirality implies chemical chirality, chemical chirality implies geometrical chirality, and each inverse does not hold. We refer the readers to [26,27] for details. The purpose of this paper is to consider topological chirality of double crossover DNA polyhedral links and 3-regular protein polyhedral links. We shall ignore both the DNA and protein sequence and use the orientation of the two backbone strands of the dsDNA to orient DNA polyhedral links. The term "dsDNA" is defined as the double stranded DNA. Thus we consider DNA polyhedral links as oriented links and protein polyhedral links as unoriented ones.

Many tools have been used to study the chirality of DNA and protein knots and links. In [28], the authors introduced an uni-variable polynomial from the adjacency matrix of a link diagram. The point symmetry group was used to detect the chirality in [23] and the writhe was applied to detect the topological chirality in [24,29]. The HOMFLY polynomial [30,31] is more powerful, but difficult to compute.

See [29,32–36]. The tool we shall use is the celebrated Jones polynomial [37] for oriented links and its counterpart for unoriented links. The advantage is that sometimes the Jones polynomial can deal with the chirality of alternating links with writhe 0 or self-writhe 0. The form of the Jones polynomial we shall take is Kauffman's version, formed from the writhe (or self-writhe) and the Kauffman bracket polynomial [38]. However, computing the Jones polynomial of links with large number of crossings such as double crossover DNA polyhedral links and 3-regular protein polyhedral links is also very difficult, we shall not do this. Our aim is to deduce some easily computable chirality criteria from the symmetry of Jones polynomial of achiral links.

A polyhedron P is a solid in $\mathbb{R}^3$, enclosed by a number of polygons (faces), any pair of polygons having at most one side in common, while three or more polygons having at most one vertex in common. Note that the surface of a polyhedron is topologically homeomorphic to the sphere S^2. Thus the graph consisting of vertices and edges of a polyhedron, i.e the 1-skeleton, is a planar graph via the well-known stereographic projection [39]. Accordingly, DNA and protein polyhedral links in three-dimensional space can be deformed into a plane to become so-called link diagrams. It is well known that there is an one-to-one correspondence between link diagrams and signed plane graphs [40–42].

In this paper we observe that link diagrams corresponding to bipartite plane graphs have antiparallel orientations; the absolute value of its writhe is equal to the number of edges of the bipartite plane graph. Since the writhe is not zero, the link corresponding to a bipartite graph with antiparallel orientation is chiral. As an application, the double crossover DNA polyhedral links are topologically chiral. As for the 3-regular polyhedral links, its self-writhe is 0, so the self-writhe chirality detector is invalid. We derive some simple chirality criteria for unoriented links with 0 self-writhe from the Jones polynomial. Using these criteria, we show that 3-regular protein polyhedral links are also topologically chiral. In addition, we prove that link diagrams corresponding to even, bipartite, plane graphs always have self-writhe 0.

2 Preliminaries

In this section, we give preliminary results that are all well known for knot theorists in the field of combinatorial knot theory. These results are necessary for us to give strict chirality criteria in Sect. 3.

2.1 Basic concepts, terminology and notations

A *graph G* is a pair of sets $V(G)$ and $E(G)$, where $V(G)$ is a non-empty finite set (of *vertices*) and $E(G)$ is a multi-set of unordered pairs (x, y) (not necessarily distinct) of vertices called *edges*. An edge with unordered pair (x, x) is called a *loop*. A graph G is said to be *connected* if, for any two distinct vertices $u, v \in V(G)$, there is a path $u = u_0 u_1 u_2 \cdots u_l = v$, where u_i $(i = 0, 1, \ldots, l)$ are all distinct and (u_{i-1}, u_i) is an edge for $i = 1, 2, \ldots, l$. A *component* of a graph G is a subgraph that is connected and is not properly contained in any other connected subgraph of G. A *bridge* of a graph G is an edge whose removal would increase the number of connected components of

G. A graph is said to be *trivial* if it consists of only an isolated vertex without loops. A graph is called *reduced* if it is connected, loopless and bridgeless.

A graph is *planar* if it can be embedded in the plane; that is, it can be drawn on the plane so that no two edges intersect. A *plane* graph is a particular planar embedding of a planar graph. We use $v(G)$ to denote the number of vertices, $e(G)$ the number of edges, $f(G)$ the number of faces, respectively, of a plane graph G. A *signed* graph is a graph each of whose edges is labeled with a sign ($+$ or $-$). A signed graph is called *positive* (resp. *negative*) if each of its edge receives a positive (resp. negative) sign.

A graph $G = (V, E)$ is said to be *bipartite* if V can be divided into two nonempty subsets V_1 and V_2 with $V_1 \cup V_2 = V$ and $V_1 \cap V_2 = \emptyset$ such that for every edge $e \in E$, one end-vertex of e belongs to V_1 and the other end-vertex of e belongs to V_2. It is well known that a graph G is bipartite if and only if G does not contain a circuit of odd length [39]. A graph G is said to be *even* if each of its vertices has even degree. A connected even graph is the so-called Eulerian in [39]. Let G be a connected even plane graph. The *planar dual* of a plane graph G is a graph that has a vertex corresponding to each face of G, and an edge joining two neighboring faces for each edge in G. We shall denote by G^* the planar dual of G. There is a natural bijection between edges of G and edges of G^*. When G is signed, G^* will also be signed but with the sign of each corresponding edge reversed. The following lemma is a folklore [see Excise 9.2.3 (b) of [39]], for the proof, please see [43].

Lemma 2.1 *Let G be a connected plane graph. Then G is even if and only if G^* is bipartite.*

A *knot* is a simple closed piecewise linear curve in Euclidean 3-space $\mathbb{R}^3$. A *link* is the disjoint union of finite number of knots; each knot is called a *component* of the link. We take the convention that a knot is a one-component link. We can always represent links in $\mathbb{R}^3$ by *link diagrams* in a plane, that is, regular projections with a short segment of the underpass curve cut at each double point of the projection. A link diagram is said to be *alternating* if over- and under-crossings alternate as one travels the link (crossing at the crossings). A link is said to be *alternating* if it has an alternating link diagram. A *nugatory* crossing of a link diagram is a crossing in the diagram so that two of the four local regions at the crossing are part of the same region in the larger diagram. A *reduced* diagram is one that is connected and does not contain nugatory crossings.

2.2 Links and graphs

The 1–1 correspondence between link diagrams and signed plane graphs has been known for more than one hundred years. It was one of the methods used by Tait and Little in the late nineteenth century to construct a table of knot diagrams of all knots starting with graphs with a relatively small number of edges and then increasing the number of edges [40]. To describe this correspondence, we first recall the medial graph of a plane graph.

The *medial graph* $M(G)$ of a non-trivial connected plane graph G is a 4-regular plane graph obtained by inserting a vertex on every edge of G, and joining two new

Fig. 1 The correspondence
between a crossing and a signed
edge
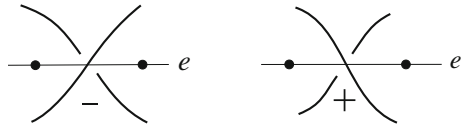
vertices by an edge lying in a face of G if the vertices are on adjacent edges of the face; if G is trivial, its medial graph is defined to be a simple closed curve surrounding the vertex (strictly, it is not a graph); if a plane graph G is not connected, its medial graph $M(G)$ is defined to be the disjoint union of the medial graphs of all its connected components.

Given a signed plane graph G, we first draw its medial graph $M(G)$. To turn $M(G)$ into a link diagram $D(G)$, we turn the vertices of $M(G)$ (i.e., edges of G) into crossings by defining a crossing to be over or under according to the sign of the edge as shown in Fig. 1. Conversely, given a connected link diagram D, shade it as in a checkerboard so that the unbounded face is unshaded. Note that such a shading is always possible, since link diagrams can be viewed as 4-degree plane graphs, see Exercise 9.6.1 ofvBondy. We then associate D with a signed plane graph $G(D)$ as follows: For each shaded face F, take a vertex v_F, and for each crossing at which F_1 and F_2 meet, take an edge (v_{F_1}, v_{F_2}) and give the edge a sign also as shown in Fig. 1. If a link diagram D is not connected, its corresponding signed plane graph $G(D)$ is defined to be the disjoint union of the signed plane graphs of all its connected components.

Under the 1–1 correspondence described above, there is also a 1–1 correspondence between edges of G and crossings of $D(G)$. Note that $D(G)$ is alternating if and only if G is positive or negative. If we take the sphere S^2 for the plane, then $D(G) = D(G^*)$.

2.3 Jones and Tutte polynomials

The Kauffman bracket polynomial $\langle D \rangle$ of an unoriented link diagram D was introduced by Kauffman in [38]. See also [44,45]. It not only provides a simple state model for the Jones polynomial [37] but also connects knot theory to statistical mechanics [46]. The Kauffman square bracket polynomial $[D] = [D](A, B, d) \in \mathbb{Z}[A, B, d]$ of a link diagram D can be defined by the following two rules:

1. The Kauffman square bracket polynomial of a diagram consisting of n disjoint simple closed curves in the plane is d^{n-1};
2. For any crossing c of the link diagram D, $[D] = A[D_c^A] + B[D_c^B]$, where D_c^A and D_c^B are link diagrams obtained from D by opening the A-channel and B-channel of the crossing c, respectively (see Fig. 2).

The Kauffman bracket polynomial $\langle D \rangle \in \mathbb{Z}[A, A^{-1}]$ of a link diagram D is related to the Kauffman square bracket polynomial $[D]$ by

$$\langle D \rangle = [D](A, A^{-1}, -A^2 - A^{-2}). \tag{1}$$

Let D be an oriented link diagram. The writhe $w(D)$ [resp. self-writhe $s(D)$] of D is defined to be the sum of signs of the crossings (resp. self-crossings) of D. See Fig. 3
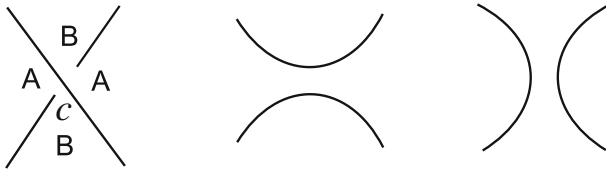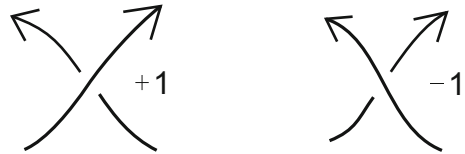
Fig. 2 A crossing c of a diagram D (*left*), D_c^A (*center*) and D_c^B (*right*) obtained from D by opening the A-channel and B-channel of the crossing c

Fig. 3 Rules for signs of crossings of an oriented link diagram

for rules of signs of crossings and Fig. 11 for examples of self-crossings. Note that the sign of a self-crossing is actually independent of the orientation of the link diagram.

The relation between the Jones polynomial and Kauffman bracket polynomial is represented as follows.

Theorem 2.2 [38,41,44]

(1) *Let D be an oriented link diagram. Let $F(D)(A) = (-A^3)^{-w(D)}\langle D \rangle$. Then the polynomial $F(D)$ in the variable A is an invariant of ambient isotopy and*

$$V_D(t) = F(D)(t^{-1/4}),$$

where $V_D(t)$ is the Jones polynomial of the link D represents.

(2) *Let D be an unoriented link diagram. Let $U(D) = (-A^3)^{-s(D)}\langle D \rangle$. Then the polynomial $U(D)$ in the variable A is an invariant of ambient isotopy.*

It is well known that the writhe (resp. self-writhe) computed from a reduced alternating link diagram is a chirality detector. However, there exist many chiral links with diagrams having writhe (resp. self-writhe) 0. For such links, the polynomials $F(D)$ and $U(D)$ may have the ability to detect their chirality. We have

Theorem 2.3 [38,41,44]

(1) *If an oriented link D is achiral, then $F(D)(A) = F(D)(A^{-1})$. This implies that if the sum of the maximum degree and the minimum degree of the $\langle D \rangle$ is not equal to $6w(D)$, then the oriented link D is chiral.*

(2) *If an unoriented link D is achiral, then $U(D)(A) = U(D)(A^{-1})$. This implies that if the sum of the maximum degree and the minimum degree of $\langle D \rangle$ is not equal to $6s(D)$, then the unoriented link D is chiral.*

Based on the correspondence between link diagrams and signed plane graphs, the Kauffman bracket polynomial was converted to the Tutte polynomial of signed graphs by Kauffman [42,44]. Let G be a signed graph. We denote by $T(G)$ the Tutte polynomial of G. When G is a signed plane graph, the Tutte polynomial of G equals exactly the Kauffman bracket of the the corresponding $D(G)$, that is

Fig. 4 Antiparallel and parallel orientations
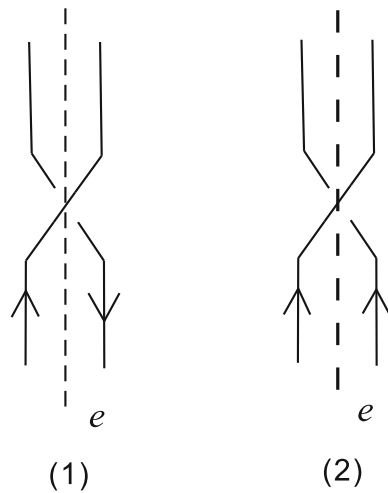
Theorem 2.4 [42,44] *Let G be a signed plane graph, D(G) be the corresponding link diagram. Then*

$$T(G) = \langle D(G) \rangle.$$

Theorem 2.5 [47,48] *Let G be a reduced positive graph. Let $v = v(G), e = e(G)$ and $f = f(G)$. Then the highest and lowest degrees of $T(G)$ are $3e - 2v + 2$ and $-e - 2v + 2$, respectively.*

3 Our chiral criteria

In this section, we provide several mathematical criteria for determining the chirality of oriented and unoriented links.

3.1 Oriented case

This subsection concerns DNA polyhedral links. The following lemma is no more than an observation.

Lemma 3.1 *Let G be a connected positive (resp. negative) plane graph. Then*

(1) *G is bipartite $\Longleftrightarrow$ D(G) has an antiparallel orientation as shown in* Fig. 4 (1). *Under this orientation, $w(D(G)) = -e(G)$ [resp. $w(D(G)) = e(G)$].*
(2) *G is even $\Longleftrightarrow$ D(G) has a parallel orientation as shown in* Fig. 4 (2). *Under this orientation, $w(D(G)) = e(G)$ [resp. $w(D(G)) = -e(G)$].*

Proof By Lemma 2.1 and Fig. 5, it suffices to prove (1). If *G* is bipartite, then the vertices of *G* can be divided into two sets *M* and *N* (see Fig. 6, left) such that each

Fig. 5 By taking the dual of G, antiparallel orientation of G becomes parallel orientation of G^*
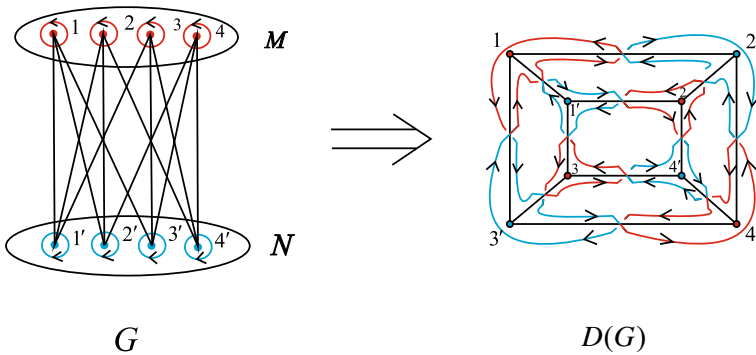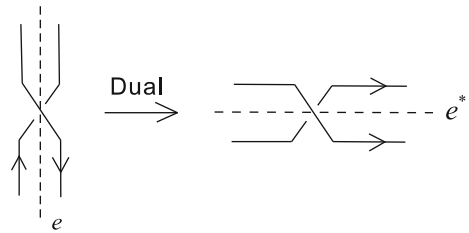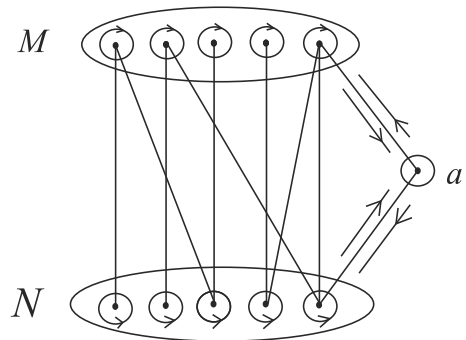

Fig. 6 A bipartite graph G produces a link $D(G)$ with an antiparallel orientation

Fig. 7 The link corresponding to a non-bipartite graph G has no antiparallel orientations

edge has an end-vertex in M, the other in N. Assign an anticlockwise (resp. clockwise) orientation to every vertex in M (resp. N), so that the link travels around the vertex in this direction. Then we obtain an antiparallel orientation of $D(G)$ as shown in the right of Fig. 6. If G is not bipartite, then $D(G)$ will have no antiparallel orientations, as shown in Fig. 7, because the orientation of the cycle around the vertex a can not be selected. As shown in Fig. 4 (1) (resp. (2)), if G is positive, then each crossing of $D(G)$ with antiparallel (resp. parallel) orientation is negative (resp. positive). Hence, if G is positive, we have $w(D(G)) = e(G)$ when G is even and $w(D(G)) = -e(G)$ when G is bipartite. The case for the negative G can be dealt with similarly. □

Theorem 3.2 *Let G be a reduced non-trivial plane graph, positive or negative. Then*

(1) *If G is bipartite, then the link $D(G)$ with the antiparallel orientation is chiral.*

(2) *If G is even, then the link D(G) with the parallel orientation is also chiral.*

Proof The writhe of a reduced alternating oriented diagram is an invariant of the oriented link it represents [27,49,50]. Thus, if the writhe of a connected reduced alternating oriented diagram is different from zero, then the oriented link it represents is chiral. Theorem 3.2 follows directly from Lemma 3.1.   □

3.2 Unoriented case

This subsection concerns protein and molecular links that are usually considered to be unoriented.

In general the self-writhe of an unoriented link diagram is not easy to determine. For a special family, we shall prove that they have self-writhe 0. Let G be a plane graph. Let $d(v)$ (resp. $d(f)$) be the degree of the vertex v (resp. the face f) of G. We denote by v_i (resp. f_i) the number of vertices (resp. faces) of degree i.

Lemma 3.3 *Let G be a connected, even, bipartite, non-trivial plane graph. Then $v_2 + f_2 \geq 4$.*

Proof Let v, e and f be the number of vertices, edges and faces of G, respectively. Recall that a non-trivial bipartite graph has no circuits of odd length. Then we have

$$v = v_2 + v_4 + v_6 + v_8 + \cdots$$
$$f = f_2 + f_4 + f_6 + f_8 + \cdots .$$
$$e = \frac{1}{2} \sum_{v \in V} d(v) = v_2 + 2v_4 + 3v_6 + 4v_8 + \cdots$$
$$e = \frac{1}{2} \sum_{f \in F} d(f) = f_2 + 2f_4 + 3f_6 + 4f_8 + \cdots ,$$

where F is the set of faces (including the unbounded face) of G. By Euler's formula [39], we have

$$\begin{aligned}
4 &= 2v + 2f - 2e \\
&= 2(v_2 + v_4 + v_6 + v_8 + \cdots) + 2(f_2 + f_4 + f_6 + f_8 + \cdots) \\
&\quad -(v_2 + 2v_4 + 3v_6 + 4v_8 + \cdots) - (f_2 + 2f_4 + 3f_6 + 4f_8 + \cdots) \\
&= v_2 + f_2 - (v_6 + 2v_8 + \cdots) - (f_6 + 2f_8 + \cdots).
\end{aligned}$$

Thus the lemma holds.   □

Theorem 3.4 *Let G be a connected, even, bipartite plane graph, positive or negative. Then no crossing of $D(G)$ is a self-crossing. In particular, we have $s(D(G)) = 0$.*

Proof We shall prove this by induction on $e(G)$, the number of edges of G. If $e(G) = 0$ the theorem holds trivially since $D(G)$ has no crossings. If $e(G) = 2$, then $D(G)$ is
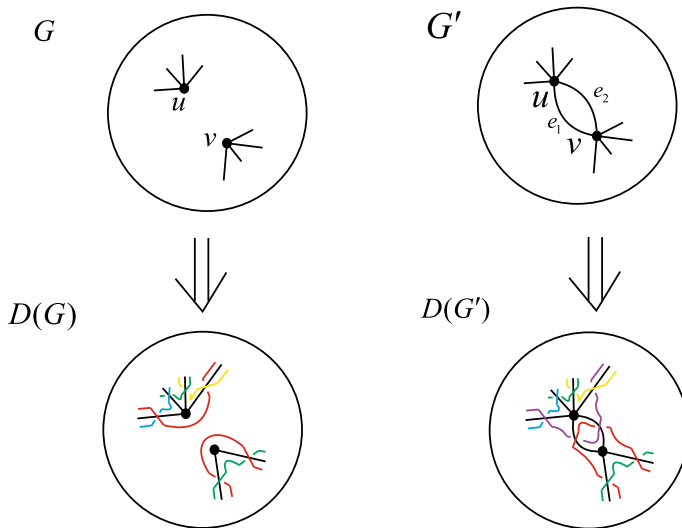
Fig. 8 G' with a face of degree 2 with boundary edges e_1, e_2 and $G = G' - e_1 - e_2$ (*up*), and their corresponding link diagrams (*down*)

the Hopf link, the theorem holds clearly. Assume as an inductive hypothesis that the theorem holds for all connected, even, bipartite plane graph with $e(G) < k$. Let G' be a connected, even, bipartite plane graph with $e(G') = k$. There are two cases.

Case 1 $f_2(G') > 0$. This means that G' has a face of degree 2 with edges e_1, e_2 as shown in Fig. 8 (right). Let $G = G' - e_1 - e_2$ as shown in Fig. 8 (left).

If G is not connected, we suppose that $G = G_1 \cup G_2$. By induction hypothesis, $D(G_1)$ and $D(G_2)$ both have no self-crossings. The two red arcs in Fig. 8 (left and down) belong to different components of the corresponding link, and hence $D(G')$ has no self-crossings. See Fig. 8 (right and down).

If G is connected, we only need to show that the two red arcs in Fig. 10 (right) belong to different components of the corresponding link, since $D(G)$ has no self-crossings by the induction hypothesis. Otherwise, suppose that the two red arcs belong to a single component of the corresponding link. Note that the component of $D(G)$ is in fact a left-right path of G (an example illustrating the notion is shown in Fig. 9), so the component must go through an even number of edges of G, since G is bipartite and hence has no odd circuits. Let $l(a \cdots c)$ and $l(b \cdots d)$ be the length of left-right path corresponding to the red component from a to c and from b to d respectively (see Fig. 10). Since G is bipartite, either $l(a \cdots c)$ and $l(b \cdots d)$ are both even or they are both odd. Note that G and its dual G^* correspond to the same diagram, i.e. $D(G) = D(G^*)$. The red component will correspond the left-right paths $a^*, \ldots, c^*, d^*, \ldots, b^*$ in G^*. Furthermore, $a^*, \ldots, c^*$ constitutes a circuit of G^*. Since G is even, we know G^* is bipartite, and hence the length of the circuit $a^*, \ldots, c^*$ is even. Thus, $l(a \cdots c)$ and $l(b \cdots d)$ are both even. Therefore, $a, \ldots, c, e_1$ will be an odd circuit of G', a contradiction.
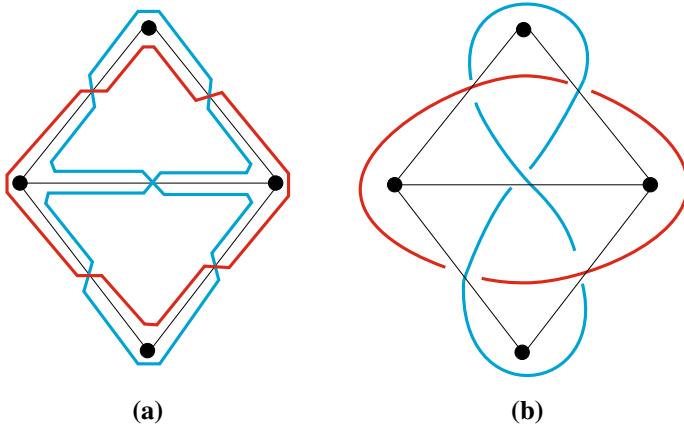
(a) **(b)**

Fig. 9 **a** Two left-right paths (*red and blue*) of a plane graph; **b** the link diagram corresponding to the plane graph which consists of two components (*red and blue*) (Color figure online)
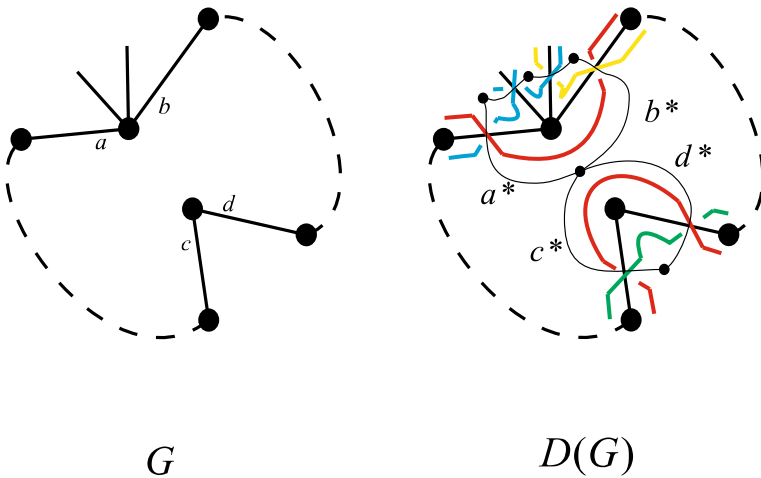

G $D(G)$

Fig. 10 G with edges labeled a, b, c, d (*left*); $D(G)$ and the dual G^* with edges labeled a^*, b^*, c^*, d^* (*right*)

Case 2. $f_2(G') = 0$.

By Lemma 3.3, $v_2(G') > 0$. Note that $D(G') = D(G'^*)$, we only need to consider the dual graph G'^* of G' which is also connected, even, bipartite plane graph, but has a face of degree 2. □

There exist non-even, bipartite plane graphs whose corresponding link diagram has self-crossings. There are also even, non-bipartite plane graphs whose corresponding link diagrams have self-crossings. Two such examples are shown in Fig. 11 and edges corresponding to self-crossings are thickened.

Theorem 3.5 *Let G be a reduced plane graph with $s(D(G)) = 0$, positive or negative. Then if $e(G) \neq 2v(G) - 2$, then the unoriented link $D(G)$ represents is chiral. In*
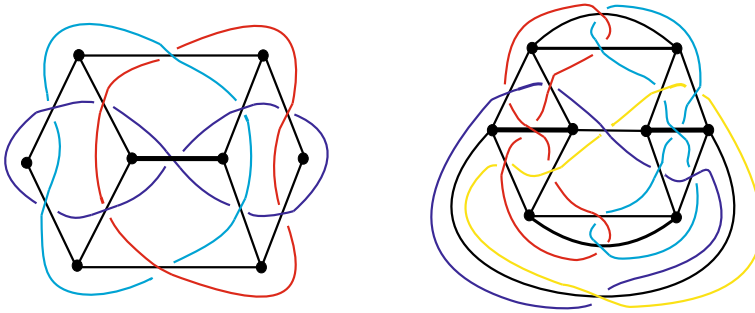
Fig. 11 A non-even, bipartite plane graph (*left*); an even, non-bipartite plane graph (*right*). Their corresponding link diagrams both have self-crossings

particular, when G is an even, bipartite, reduced plane graph with $e(G) \neq 2v(G) - 2$, the unoriented link $D(G)$ represents is chiral.

Proof The first part follows from Theorems 2.3 (2) and 2.5. The second part then follows from Theorems 3.4 and 2.5.   □

4 Analyses and results

In this section we shall show that a type of DNA polyhedral link, a type of protein polyhedral links and a type of molecular links are both topologically chiral. Note that we shall ignore protein sequence and use the orientation of the two backbone strands of dsDNA to orient DNA polyhedral links. Thus we consider DNA polyhedral links as oriented links and protein polyhedral links as unoriented ones. Note that topological chirality always implies chemical chirality. We obtain that the two types of DNA polyhedra and protein polyhedra are both (chemical) chiral. In fact, the chirality of double crossover octahedra link has been revealed by cryogenic electron microscopy (cryoEM) [15].

4.1 Double crossover DNA polyhedral links

In recent years, chemists and biologists, in laboratory, designed and synthesized some fancy double crossover DNA polyhedra, such as DNA tetrahedron [16], cube [17], octahedron [15], dodecahedron[16], icosahedron [18] and buckyball [16], by covering each vertex of degree n of the polyhedron by "n-point star motif (tiles)" and through sticky-end association between the tiles. The "n-point star motif" has an n-fold rotational symmetry and consists of $2n + 1$ single strands: a long repetitive central DNA strand (colored red and yellow), n identical medium DNA strands (colored green) and n identical short DNA strands (colored black). The colored yellow part at the center of the motif represents n unpaired DNA single-strands whose lengths can be adjusted to change bending degree of the whole structure. See Fig. 12. In Fig. 13b, using 8 "3-star points" to cover every vertex of the cube graph in Fig. 13a, the resultant links are 4-turn DNA cube and 4.5-turn DNA cube, called double crossover cubic links [17].
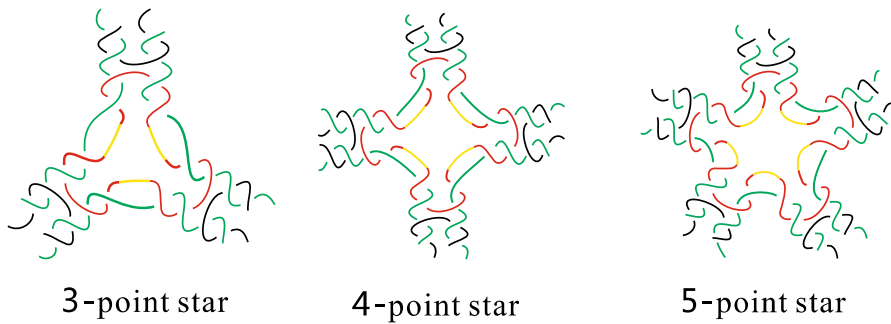
Fig. 12 3-point star, 4-point star and 5-point star

Note that along each edge, there are two anti-parallel DNA duplexes [15], thus we view double crossover polyhedral links as oriented links. See also [19,51]. Their braid indices have been determined in [52,53], which, however, are useless for detecting chirality.

In the following, we first consider the chirality of 4-turn DNA cube. In a similar way, the chirality of 4.5-turn DNA tube can also be illustrated.

In general, the plane graph G corresponding to a double crossover polyhedral link based on the polyhedron P can be obtained from the 1-skeleton of the polyhedron P by the following steps:

Step 1: Truncating the polyhedron P, i.e. cutting each corner of the polyhedron P, we obtain a polyhedral graph Q.

Step 2: Flattening edges of Q corresponding to edges of P, i.e. converting a single edge into two parallel edges, we obtain a plane graph H.

Step 3: Subdividing each edge of H by inserting three vertices, we obtain the plane graph G corresponding to the double crossover polyhedral link based on the polyhedron P.

An example is given in Fig. 14. To obtain G, every edge of H is replaced by a path of length 4, so G has no odd circuits and hence G is bipartite. According to Theorem 3.2 (1), we know that all DNA double crossover 4-turn polyhedral links are topologically chiral.

According to Ref [17], for a double crossover 4.5-turn polyhedral links $L(P)$, the polyhedron P must be a bipartite. So, in a similar way, according to Theorem 3.2 (1), DNA double crossover 4.5-turn polyhedral links are topologically chiral.

4.2 3-regular protein polyhedral links

Now we consider protein polyhedra. By now, as far as we know, only one protein polyhedra has been found, that is HK97 capsid, a topologically linked protein catenane in the mature empty capsid of the double-stranded DNA bacteriophage [22]. Motivated by this discovery, Qiu et al. [54] developed a method of constructing polyhedral links based on polyhedra by means of "three-cross curves and untwisted double-line"
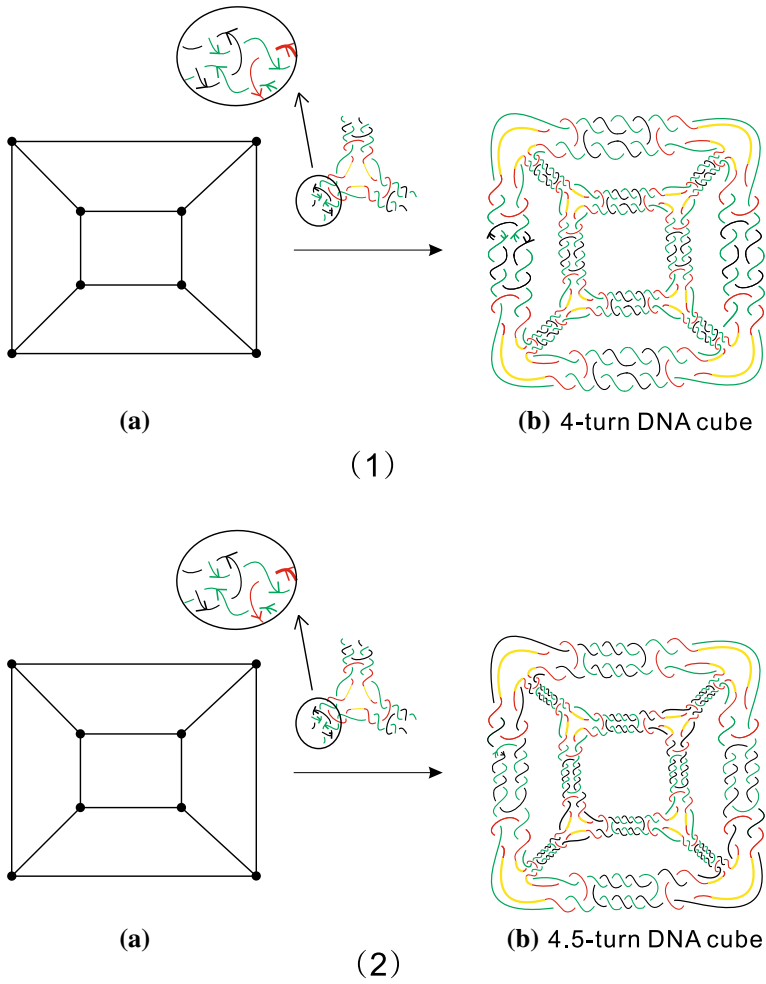
(a)   **(b)** 4-turn DNA cube

（1）

(a)   **(b)** 4.5-turn DNA cube

（2）

Fig. 13 **a** The cubic graph; **b** the double crossover DNA cubic link

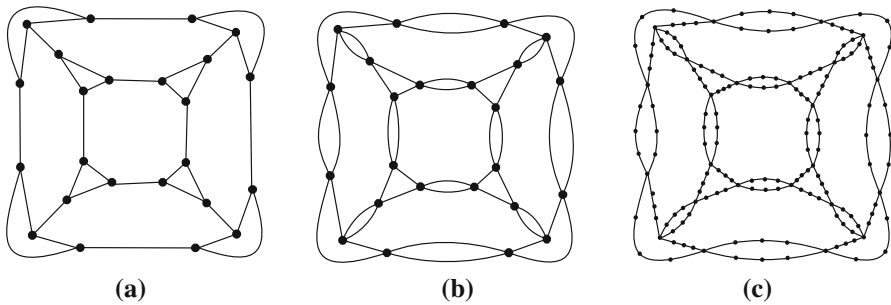**(a)**   **(b)**   **(c)**

Fig. 14 **a** The truncated cubic graph (Q); **b** its flattening (H) and **c** subdivision (G)
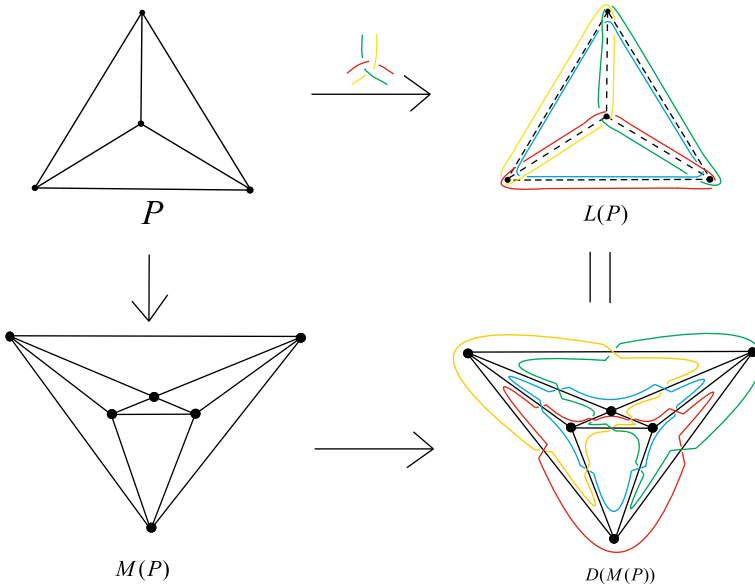
Fig. 15 The tetrahedral graph (*up* and *left*), 3-regular tetrahedral link (*right*) and its corresponding plane graph (*down* and *left*)

covering in [24]. We shall call them 3-regular protein polyhedral links. We shall ignore protein sequence and view protein polyhedral links as unoriented links.

Let P be a polyhedron of degree 3. Let $L(P)$ be the polyhedral link based on P constructed by means of "three-cross-curves" covering. See Fig. 15 for an example. We have two observations:

(1) Each face of P corresponds to a non-intersecting component of $L(P)$ [see Fig. 15 (right) for an example], hence the self-writhe of $L(P)$ is zero.
(2) The plane graph corresponding to $L(P)$ is exactly the medial graph $M(P)$ of the polyhedral graph of P (see Fig. 15), hence $v(M(P)) = e(P)$ and $e(M(P)) = \frac{4v(M(P))}{2} = 2v(M(P)) = 2e(P)$.

By Theorem 3.5, $L(P)$ is topologically chiral. Since chiral unoriented links must be also chiral as oriented links. Even if we consider protein sequence and view protein polyhedral links as oriented ones, they are still chiral.

5 Summary and discussions

In this paper, we present a criterion for oriented alternating links to be chiral; that is, if they are formed from a reduced non-trivial bipartite plane graphs and they have an antiparallel orientation, then they are topologically chiral as oriented links. This criterion is enough to deal with double crossover DNA polyhedral links with 4 turns (resp. 4.5 turns) or more generally, even turns (reps. odd turns). We noted that DNA double crossover molecules were modelled in 1993 [19]. We also present a criterion

for unoriented links with self-writhe 0; that is, if they are formed from a reduced plane graph with $e(G) \neq 2v(G) - 2$, then they are topologically chiral. This criterion is used to obtain the chirality of 3-regular protein polyhedral links. In theory, it is interesting to characterize reduced plane graphs whose corresponding link diagrams have zero self-writhes. This is not solved and we only prove the self-writhe of an unoriented link formed from a connected, even , bipartite plane graph is zero.

We considered the double crossover 4-turn and 4.5-turn DNA polyhedra in this paper. Our criterion for DNA polyhedral links can be used to deal with cycle-crossover polyhedral links introduced in [55]. In addition, as a theoretically possible synthesizing method, for any $k \geq 3$, "k-crossing curves" covering was proposed in [25,56,57] to construct protein polyhedral links. We shall study chirality of such polyhedral links in the subsequent paper.

Conflict of interest The authors declare that they have no conflict of interest.

References

1. M.A. Krasnow, A. Stasiak, S.J. Spengler, F. Dean, T. Koller et al., Nature **304**, 559 (1983)
2. D.W. Sumners, Not. AMS **42**, 528 (1995)
3. F.B. Dean, A. Stasiak, T. Koller, N.R. Cozzarelli, J. Biol. Chem. **260**, 4975 (1985)
4. S.A. Wasserman, N.R. Cozzarelli, J. Biol. Chem. **266**, 20567 (1991)
5. V. Biou, R. Dumas, C. Cohen-Addad, R. Douce, D. Job et al., EMBO J. **16**, 3405 (1997)
6. S. Misaghi, P.J. Galardy, W.J.N. Meester, H. Ovaa, H.L. Ploegh et al., J. Biol. Chem. **280**, 151 (2005)
7. http://pknot.life.nctu.edu.tw/table_new.php
8. J. Chen, N.C. Seeman, Synthesis from DNA of a molecule with the connectivity of a cube. Nature **350**, 631 (1991)
9. R.P. Goodman, R.M. Berry, A.J. Turberfield, Chem. Commun. **12**, 1372 (2004)
10. W.M. Shih, J.D. Quispe, G.F. Joyce, Nature **427**, 618 (2004)
11. Y. Zhang, N.C. Seeman, J. Am. Chem. Soc. **116**, 1661 (1994)
12. C.M. Erben, R.P. Goodman, A.J. Turberfield, J. Am. Chem. Soc. **129**, 6992 (2007)
13. J. Zimmermann, M.P. Cebulla, S. Mönninghoff, G.V. Kiedrowski, Angew. Chem. Int. Ed. **47**, 3626 (2008)
14. X. Jin, F. Zhang, MATCH Commun. Math. Comput. Chem. **65**, 501 (2011)
15. Y. He, M. Su, P. Fang, C. Zhang, A.E. Ribbe et al., Angew. Chem. Int. Ed. **49**, 748 (2010)
16. Y. He, M. Su, P. Fang, C. Zhang, A.E. Ribbe et al., Nature **452**, 198 (2008)
17. C. Zhang, S.H. Ko, M. Su, Y. Leng, A.E. Ribbe et al., J. Am. Chem. Soc. **131**, 1413 (2009)
18. C. Zhang, M. Su, Y. He, X. Zhao, P. Fang et al., PNAS **105**, 10665 (2008)
19. T.J. Fu, N.C. Seeman, Biochemistry **32**, 3211 (1993)
20. C. Lin, Y. Liu, H. Yan, Biochemistry **48**, 1663 (2009)
21. A. Kuzuya, M. Komiyama, Nanoscale **2**, 310 (2010)
22. W.R. Wikoff, L. Lilja, R.L. Duda, H. Tsuruta, R.W. Hendrix et al., Science **289**, 2129 (2000)
23. W.Y. Qiu, H.W. Xin, J. Mol. Struc. (THEOCHEM) **429**, 81 (1998)
24. Y.M. Yang, W.Y. Qiu, MATCH Commun. Math. Comput. Chem. **58**, 635 (2007)
25. X.S. Cheng, W.Y. Qiu, H. Zhang, MATCH Commun. Math. Comput. Chem. **62**, 115 (2009)
26. E. Flapan, Source: Solitons Fractals **9**, 547 (1998)
27. E. Flapan, *When Topology Meets Chemistry* (Cambridge University Press, Cambridge, 2000)
28. C. Liang, Y. Jiang, J. Theor. Biol. **158**, 231 (1992)
29. G. Hu, X.D. Zhai, D. Lu, W.Y. Qiu, J. Math. Chem. **46**, 592 (2009)
30. P. Freyd, D. Yetter, J. Hoste, W.B.R. Lickorish, K. Millett et al., Bull. Am. Math. Soc. **12**, 239 (1985)

31. J.H. Przytycki, P. Traczyk, Kobe J. Math. **4**, 115 (1987)
32. X. Jin, F. Zhang, MATCH Commun. Math. Comput. Chem. **63**, 657 (2010)
33. S.Y. Liu, H. Zhang, W.Y. Qiu, MATCH Commun. Math. Comput. Chem. **67**, 65 (2012)
34. S.Y. Liu, X.S. Cheng, H. Zhang, W.Y. Qiu, J. Math. Chem. **48**, 439 (2010)
35. X. Jin, F. Zhang, Proc. Am. Math. Soc. **140**, 1459 (2012)
36. X.S. Cheng, Y. Lei, W. Yang, J. Math. Chem. **52**, 23 (2014)
37. V.F.R. Jones, Bull. Am. Math. Soc. **12**, 103 (1985)
38. L.H. Kauffman, Topology **26**, 395 (1987)
39. J.A. Bondy, U.S.R. Murty, *Graph Theory and Its Applications* (The Macmillan press ltd., New York, 1976)
40. K. Murasugi, *Knot Theory and Its Applications* (Birkhauser Boston Inc., 1996)
41. B. Bollobás, *Modern Graph Theory* (Springer, Berlin, 1998)
42. L.H. Kauffman, Discrete Appl. Math. **25**, 105 (1989)
43. K. Murasugi, A. Stoimenow, Adv. Appl. Math. **31**, 440 (2003)
44. L.H. Kauffman, Am. Math. Monthly. **95**, 195 (1988)
45. L.H. Kauffman, *On Knots* (Annals of Mathematics Studies, Princeton University Press, Princeton, 1987)
46. L.H. Kauffman, A.M.S. Contemp, Math. Ser. **78**, 263 (1989)
47. O. Dasbach, X.S. Lin, Pac. J. Math. **231**, 279 (2007)
48. X. Jin, F. Zhang, J. Ge, When will the crossing number of an alternating link decrease by two via a crossing change? submitted
49. K. Murasugi, Math. Proc. Camb. Philos. Soc. **102**, 317 (1987)
50. K. Murasugi, Topology **26**, 187 (1987)
51. G. Hu, W.Y. Qiu, A. Ceulemans, Plos One **6**, e26308 (2011)
52. X.S. Cheng, X. Jiang, H. Dai, J. Math. Chem. **50**, 1386 (2012)
53. X.S. Cheng, X. Jin, Plos One **7**, e48968 (2012)
54. W.Y. Qiu, X.D. Zhai, J. Mol. Struct. (THEOCHEM) **756**, 163 (2005)
55. X.S. Cheng, H. Zhang, G. Hu, W.Y. Qiu, MATCH Commun. Math. Comput. Chem. **63**, 637 (2010)
56. X.S. Cheng, S.Y. Liu, H. Zhang, W.Y. Qiu, MATCH Commun. Math. Comput. Chem. **63**, 115 (2010)
57. D. Lu, G. Hu, Y.Y. Qiu, W.Y. Qiu, MATCH Commun. Math. Comput. Chem. **63**, 67 (2010)