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Marvellous molecular shapes

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ABSTRACT

A survey of a variety of interesting molecular shapes characterized mostly in the last two decades is presented. The selection of molecules shown is made based on their attractive three-dimensional structures (so often concealed by their usual two-dimensional structural formulae), the unusual coordination geometries of metal atoms, the subjective appreciation of how beautiful, funny or interesting a molecular shape is, or the sticky nature of arene rings at terphenyl groups and their effects on the molecular shapes of metal complexes.

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1. Introduction

In an inspired essay, Roald Hoffmann reflected on how chemists perceive “molecular beauty” [1]. According to Hoffmann, beauty may arise from novelty, epitomized at the time by metal clusters containing six-coordinated carbon atoms. Beauty may also be associated with molecular shape, and some examples given by him are “simple, symmetrical and devilishly hard to make”, but other forms of molecular beauty have to do with the perception of the chemist of what is unique about its composition, structure or bonding. Finally, what we may consider beautiful is the way a molecule behaves in front of some external stimulus, as exemplified by haemoglobin, a porphyrin-bearing protein that experiences important changes in response to the binding of oxygen.

In this article, I wish to present more recent examples of beautiful molecules. In some of them I find beauty in the way they surprise us by adopting a structure in space that is hard to anticipate from a structural scheme. Conversely, in some cases it is the two-dimensional (2D) scheme of a molecule that excites our imagination. The choice of

structures to be discussed is completely subjective, based on my perception of what is interesting, beautiful or just funny in the molecular shapes displayed by them.

2. Deceiving structural drawings

An example of how a structural drawing may give us poor information of a remarkable 3D molecule is provided by hexadecaphenyl-dodecabenzene [2]. Although it is drawn on paper as a square molecule (Fig. 1a), its structure in space is that of a tetrahedron such that chains of four phenyl groups occupy four out of the six edges of the tetrahedron (Fig. 1b). Continuous shape measures [3] of the centres of the four phenyl groups at the corners indicate a nearly perfect tetrahedron, with just a slight (9%) distortion towards a square. The shapes of the 2D scheme and the real molecular structure can hardly be more different!

To the variety of molecules that have the shape of daily live objects [4], two cases have been chosen that also stress the inability of 2D schemes to suggest their 3D structures. Both are fully aromatic molecules composed of fused phenyl rings around a central five- or seven-membered ring (Fig. 2a and b, respectively) that look quite similar when represented as a 2D scheme. However, their 3D

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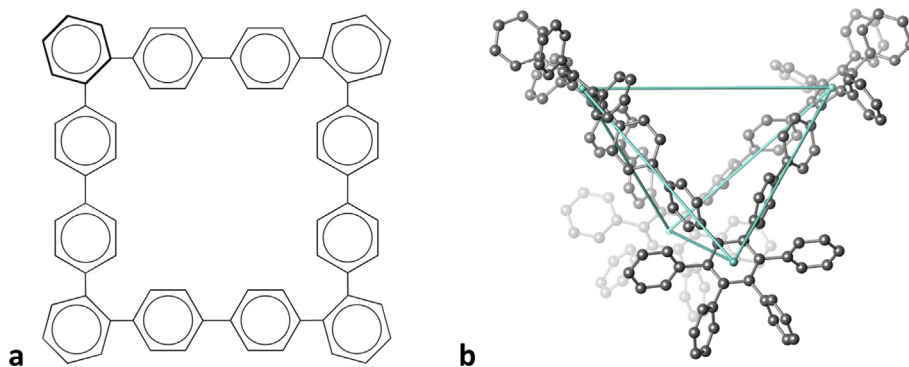


Fig. 1. Schematic drawing of a dodecabenzene skeleton (a) and its 3D tetrahedral structure (b).

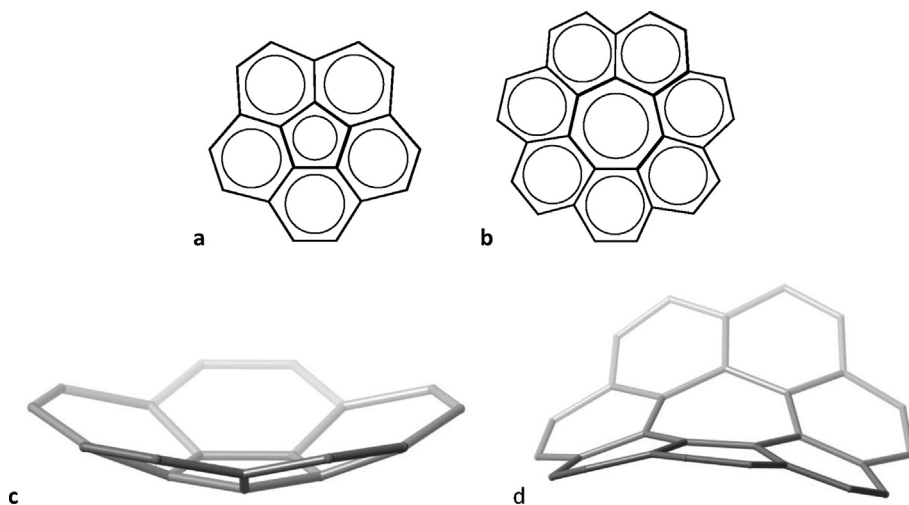


Fig. 2. Two-dimensional representation of corannulene (a) and [7]circulene (b), and their 3D structures (c and d, respectively).

structures are non-planar and very different: while the former has the shape (Fig. 2c) of a bowl [5] – and is often referred to as a “buckybowl” –, the latter is non-planar as well, but bent in such a way as to form a sawhorse [6] (Fig. 2d).

3. Impossible structural drawings

When we try to draw a sequence of fused phenyl rings everything seems to work right for up to five rings (Fig. 3a). However, our drawing suggests a planar molecule, far from what we find in its crystal structure [7], that shows a helical geometry in keeping with its name “pentahelicene” (Fig. 3b). If we wish to add one more phenyl ring, with all of the rings being regular hexagons, we end up (Fig. 3c) with a representation of coronene, $C_{24}H_{12}$, instead of hexahelicene, $C_{26}H_{16}$. To make a 2D representation of the latter, we must make use of irregular hexagons (e.g., Fig. 3d), but we still misrepresent the helical shape of the molecule. The situation worsens as we go to higher helicenes, requiring highly distorted hexagons. An alternative is to use hexagons of increasing size, to avoid the rings to overlap and additionally providing some

sense of perspective that transmits better the idea of helicity, as shown in Fig. 3e for heptahelicene. But, regardless of how we represent them in two dimensions, the 3D structures of the helicenes look highly attractive and reminiscent of the spiral staircase of the entrance Hall of the Louvre museum, as illustrated by undecahelicene [8] and hexadecahelicene [9] (Fig. 4).

4. Unusual coordination geometries

In the past years, we have extensively studied polyhedral molecular structures and their symmetry properties [10–14]. A number of supposedly uncoordinated or monocoordinated transition metal compounds were also recently analysed and showed the existence of hidden beautiful molecular shapes [15]. But the ability of chemists to design and synthesize molecules with new shapes gives me the opportunity to call the attention to some interesting new polyhedral shapes.

The coordination of a cyclodimethylsiloxane with an O_6 donor set to an $Ag(I)$ ion [16] produced a complex with three short Ag–O distances of about 2.42 Å, which would correspond to a canonical trigonal planar coordination of a

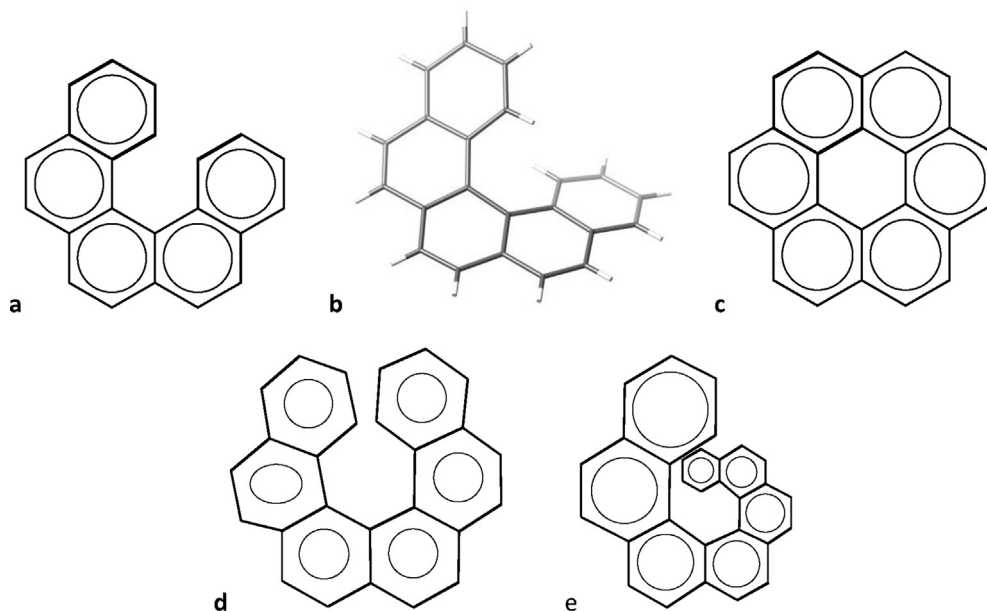


Fig. 3. Two-dimensional representation of pentahelicene (a) and its 3D molecular structure (b), an arrangement of six regular hexagons that corresponds to the formula of coronene (c), a set of six distorted hexagons representing the molecular formula of hexahelicene (d), and fused six-membered rings of different sizes that give some sense of the helical nature of heptahelicene (e).

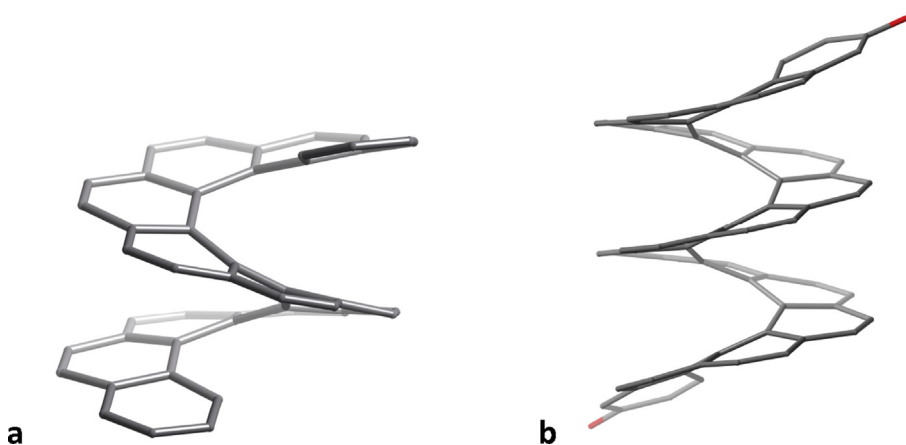


Fig. 4. Helical structures of the skeletons of undecahelicene (a) and 3,34-bis(triisopropyl-silyloxy)hexadecahelicene (b), the two terminal triisopropylsilyl groups are omitted for clarity).

d^{10} ion, but the three remaining oxygen atoms are held at not much longer distances of about 2.67 Å, thus making this molecule a reasonable example of an unusual hexagonal planar coordination of a metal ion (Fig. 5a). In a related compound the counter ion establishes a coordination bond to Ag at an axial position, thereby forming an also unusual hexagonal pyramidal coordination. A similar situation can be found [17] with the 18-crown-6 ligand that, together with an oxygen atom of the triflate counter ion, forms an apparent heptacoordination around a Ag(I) ion, in a rather distorted hexagonal pyramid (Fig. 5b).

Very high coordination numbers are rare, but once they are achieved, some unexpected coordination polyhedra can

be found, such as the octagonal antiprismatic “drum” of the CoB_{16} cluster [18] (Fig. 6a), or the hexacapped hexagonal prism of the LaNi_{18} group (Fig. 6b) in the solid state structure of LaNi_5 [19], with the so-called CaCu_5 structure found in hundreds of binary intermetallic compounds.

An attractive double decker sandwich structure (Fig. 7) combines three slices of “bread”, the outer ones formed by two different porphyrins and the inner one by a phthalocyanine ring [20]. The metal ions sandwiched by these rings are Ce(III) and Gd(III). Although both metal ions are octacoordinated, Ce is in a cubic environment and Gd in a square antiprismatic one and both polyhedra share a square face.

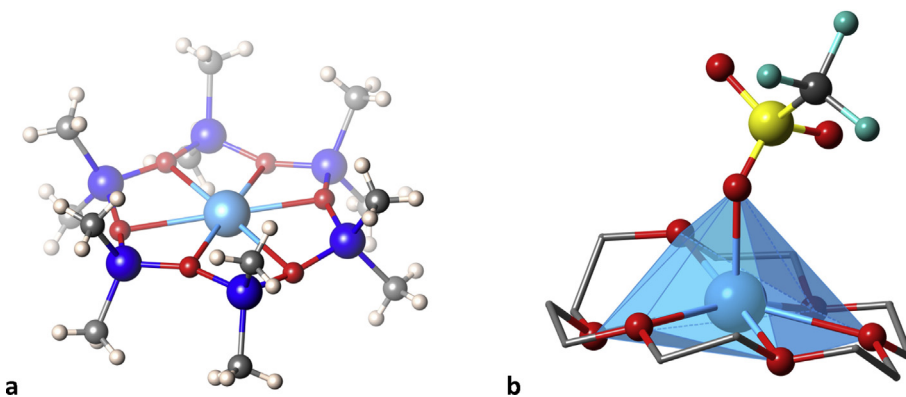


Fig. 5. Two cyclic ligands with O_6 donor sets that favour apparent hexagonal planar (a) and hexagonal pyramidal (b) coordinations to $Ag(I)$ ions.

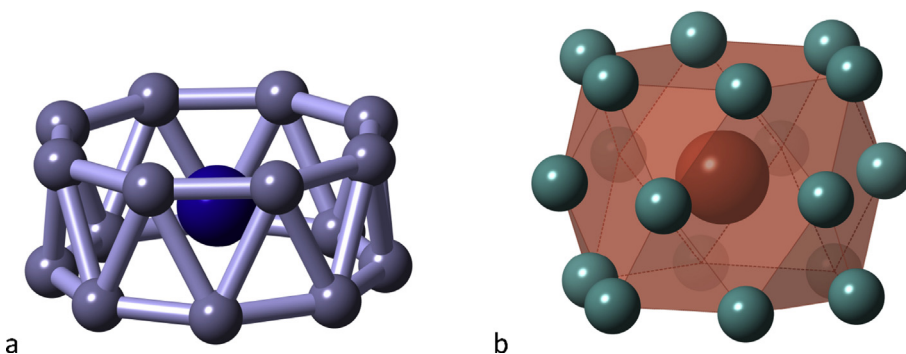


Fig. 6. Two drums with very high coordination numbers: the CoB_{16} anion (a) and the $LaNi_{18}$ group in $LaNi_5$ (b).

5. Stars and planets of several sorts

Some molecules may remind us of planets and stars. For instance, a ring made up of 11 phenyl rings surrounding a

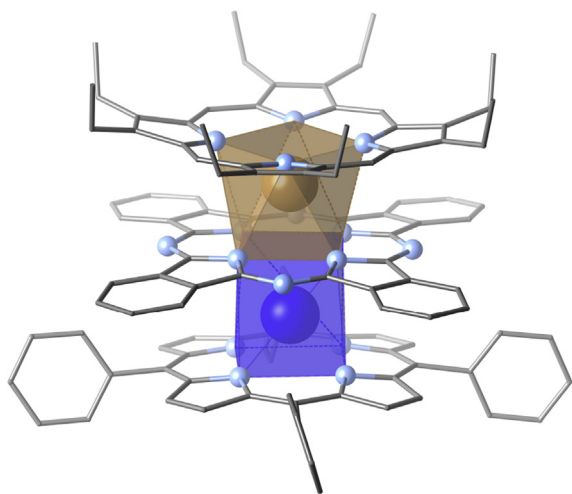


Fig. 7. A Gd and Ce double decker sandwich complex with a square antiprismatic and a cubic coordination spheres, respectively, that share a square face.

C_{70} fullerene [21], resembles the planet Saturn (Fig. 8a) with its planetary rings. Also simple molecules, such as the XeF_5^- anion [22] or the well-known tetramethylsilane, $SiMe_4$ [23], appropriately coloured, look like shining stars (Fig. 8b and c, respectively).

Some time ago Michel Verdaguer suggested me the analogy between the representation made by artist Joan Miró of stars, be them six- or eight-pointed, and the way coordination chemists schematically draw an octahedron. Some reflections on representation in art and chemistry, based on the discussions we had, can be found in a recent book [24], where an outstanding heptanuclear compound [25] created by the Jussieu group illustrates a sort of constellation formed by a central star surrounded by six more six-pointed stars (Fig. 9). It corresponds in fact to a central chromium atom connected via cyanide bridges to six nickel atoms.

6. Molecules that are just beautiful, funny or curious

Let me show here two examples of polyhedra surrounded by planar rings. One is a Pd_{13} -centred cuboctahedron with its square faces covered by $C_7H_7^+$ tropylium ions whose centres form an octahedron [26] (Fig. 10a). Related structures showcase a tetrahedron of tropylium ions, each on top of a vertex of a tetrahedral Mo_4 cluster [27]. In contrast, with the larger COT ($C_8H_8^{2-}$) ligands one

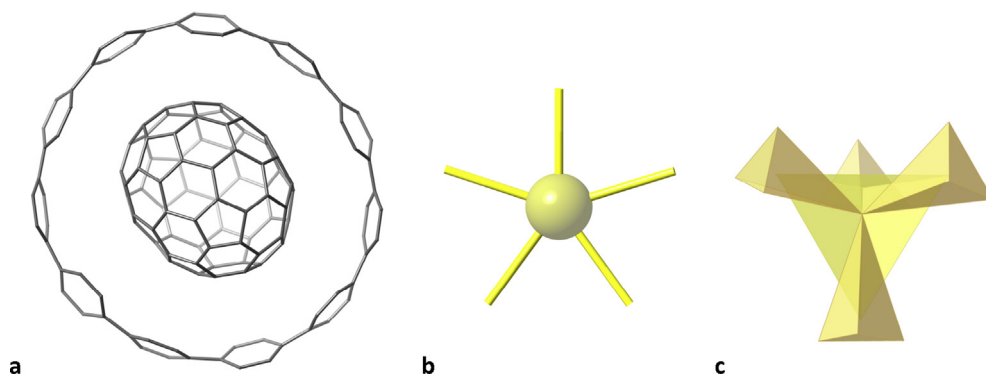


Fig. 8. A ring made up of 11 phenyl rings encircling a C_{70} fullerene that reminds the planet Saturn (a), a XeF_5 star (b) and the tetramethylsilane molecule (c).

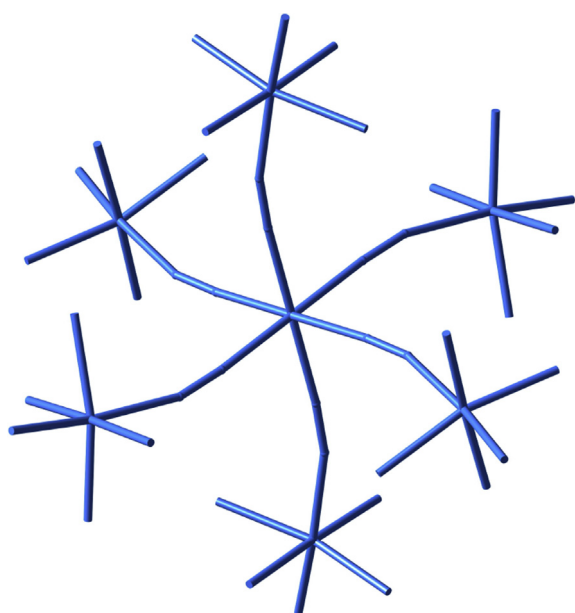


Fig. 9. A cyano-bridged heptanuclear $CrNi_6$ complex as a constellation of six-pointed stars.

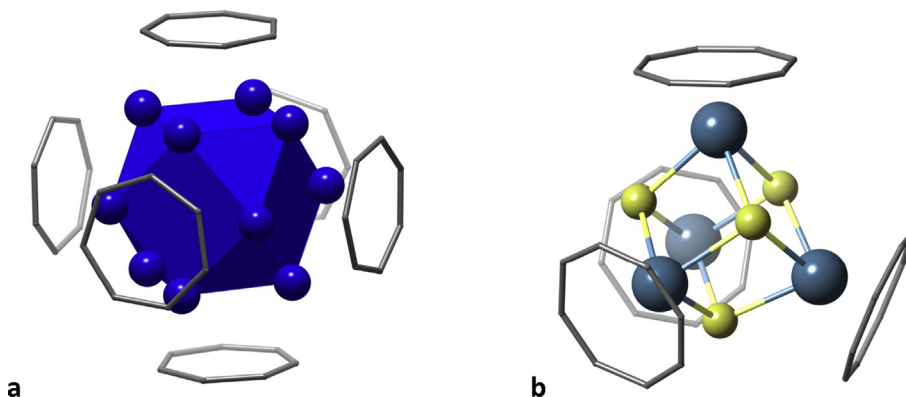


Fig. 10. Large aromatic rings covering metal clusters: (a) tropylium ions on a Pd_{13} cluster and (b) COT^{2-} anions on a Ti_4Cl_4 cubane.

can get at most four tetrahedrally arranged rings around the titanium atoms of a Ti_4Cl_4 cubane (Fig. 10b) [28].

Some molecules that are highly interesting for their architecture and bonding can also be associated with funny or curious shapes. Three examples are shown here. The extensive double bond systems of two β -carotene molecules, which are responsible for the orange colour of carrots, have been coordinated to a Pd_{10}^{2+} chain [29], altogether presenting the aspect of a hot dog sandwich (Fig. 11a). A hamburger sandwich, on the other hand, has a molecular representation in the $Pd_4(COT)_2^+$ cation [30] (Fig. 11b).

Our-third example is an organometallic Au(I) complex coordinated by two cyclopropenyl groups [31]. The $N(i-Pr)_2$ groups hanging from the cyclopropenyl rings make the ligands look like two frogs, and they face each other to kiss at the metal atom, indeed a golden kiss (Fig. 12).

7. Sticky arene rings

One of the design tools extensively developed in the past years has been the use of terphenyl groups forming a metal–carbon bond to the central ring, in such a way that the flanking arenes provide steric protection to the metal, enhanced by substituents R and R' that are commonly Me, *i*-Pr or *t*-Bu (Fig. 13a). A relevant success of this strategy is showcased by the first synthesis of a quintuple bond

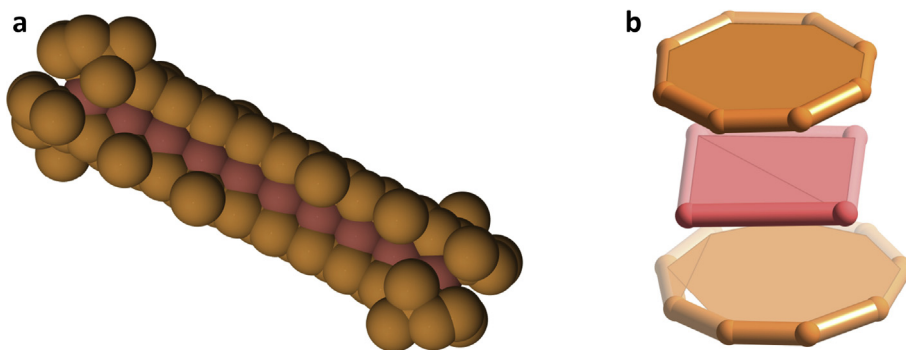


Fig. 11. Two molecular sandwiches: a hot dog sandwich (a) and a hamburger sandwich (b) in which the meat is made of palladium atoms.

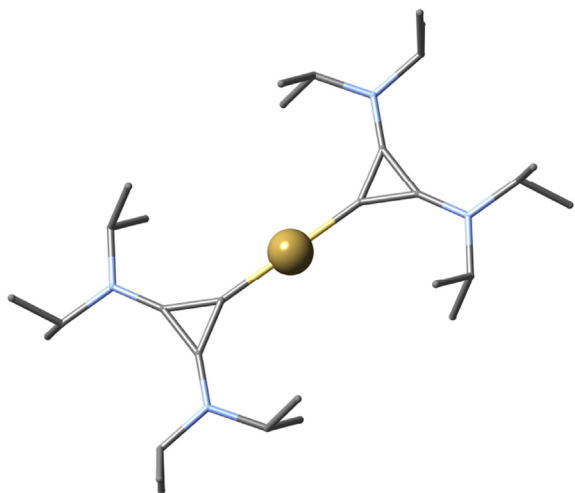


Fig. 12. A bis(cyclopropenyl)Au(I) complex representing two kissing frogs.

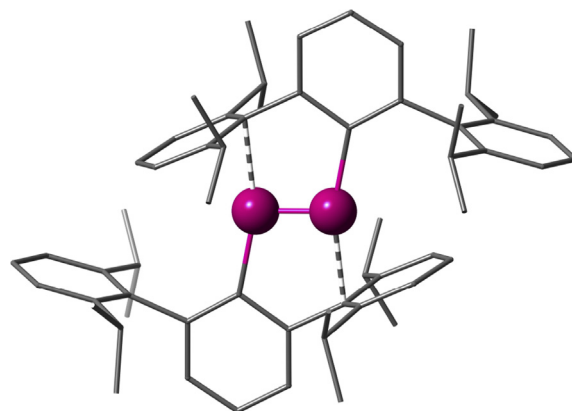


Fig. 14. Crystal structure of the first reported compound with a quintuple Cr-Cr bond [32].

between two chromium atoms, obtained by Nguyen et al. [32], subtended by only two such terphenyl groups (scheme in Fig. 13b and crystal structure in Fig. 14). Although initially the role of the flanking arene rings was assumed to be only to provide steric protection, it is now clear that besides preventing the approach of further ligands to the metal centre, they are actually π -coordinated to a metal atom in both quadruple Mo-Mo and quintuple Cr-Cr bonds [33,34], a phenomenon often referred to as “secondary interactions”.

A further step in this direction has been the addition of a coordinating group such as amido [41,43,44] or thiolato [35] to the central phenyl ring, in such a way that the terphenyl group is somewhat separated from the metal atom whereas at the same time moving it laterally (Fig. 13c).

In all of the complexes with those ligands, the flanking arene rings act as sticky flaps that try to compensate the electronic unsaturation of nearby metal centres through π -coordination with varying hapticities, which are apparently affected by the nature of the R and R' substituents. All of the structures can be classified in one of the following three cases (Table 1): (1) when both R and R' are *i*-Pr groups, the

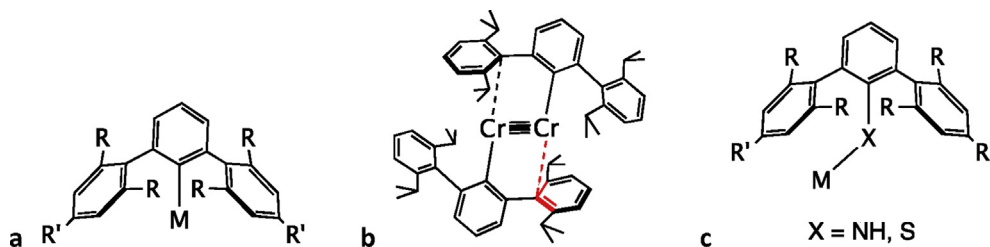


Fig. 13. Ligands with a terphenyl group that form a carbon–metal bond to a single metal centre (a) or to a multiply bonded metal (b) and related amido- and thiolato-terphenyl ligands (c).

Table 1

Geometry of the MX_2 core of bis(terphenylthiolato) and bis(terphenylamido) ($\text{X} = \text{S}$ and NH , respectively) complexes and hapticity of the flanking arene rings, together with the ranges of close interatomic $\text{M}\cdots\text{C}$ distances.

Stereochem	M	R	R'	X-M-X ($^\circ$)	Hapticity	$\text{M}\cdots\text{C}$ (\AA)	Σcov^a	Refcode	Reference
X = S									
a	Cr	<i>i</i> -Pr	<i>i</i> -Pr	180	2×1	2.50	2.15	jarjoc	[35]
a	Mn	<i>i</i> -Pr	<i>i</i> -Pr	180	2×3	2.95–3.33	2.37	jarjui	[35]
a	Co	<i>i</i> -Pr	<i>i</i> -Pr	180	2×1	2.66	2.26	jarket	[35]
a	Ni	<i>i</i> -Pr	<i>i</i> -Pr	174	2×1	2.64–2.66	2.00	jarkix	[35]
a	Zn	<i>i</i> -Pr	<i>i</i> -Pr	180	2×1	2.80	1.98	jarkod	[35]
a	Zn	<i>i</i> -Pr	H	180	2×3	2.96–3.28	1.98	lextoa	[36]
b	Fe	Me	H	117	2×2	2.46–2.92	2.28	villuu	[37]
b	Fe	Me	Me	122	2×3	2.47–2.87	2.28	piyzec	[38]
b	Zn	Me	Me	152	2×3	2.65–3.06	1.98	ligbae	[39]
c	Fe	<i>i</i> -Pr	H	120	1×6	2.48–2.61	2.28	lexsuf	[36]
c	Co	<i>i</i> -Pr	H	92	1×6	2.08–2.19	2.26	lextam	[36]
c	Ni	Me	Me	95	1×6	2.07–2.30	2.00	foqxit	[40]
X = NH									
a	Cr	<i>i</i> -Pr	H	180	2×1	2.63	2.15	sebqib	[41]
a	Cr	<i>i</i> -Pr	<i>i</i> -Pr	180	2×1	2.48	2.15	sebqex	[41]
a	Fe	<i>i</i> -Pr	<i>i</i> -Pr	180	2×2	2.79–2.94	2.28	wulweg	[42]
b	Co	Me	Me	144	2×1	2.56–2.57	2.26	sebwhi	[41]
b	Ni	Me	Me	155	2×1	2.56–2.57	2.00	sebwon	[41]
b	Mn	Me	Me	138	2, 3	2.60–2.87	2.37	wugcad	[43]
b	Fe	Me	Me	141	2, 3	2.59–2.90	2.28	wulwac	[42]

^a Sum of covalent radii of the high-spin metal atom and a carbon atom.

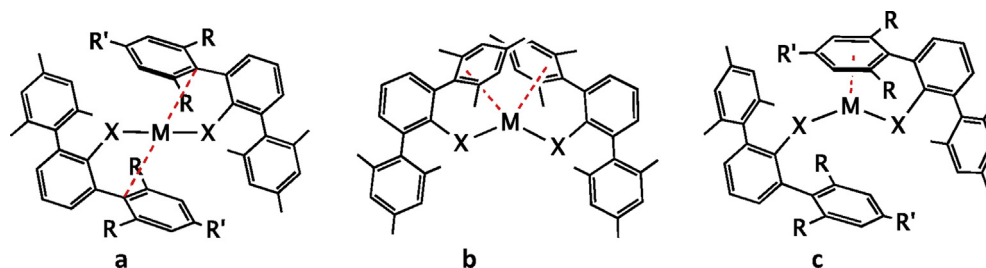


Fig. 15. Different stereochemistries of formally dicoordinated transition metal atoms with two amido- (a and b) or thiolato-terphenyl (c) ligands.

MX_2 core is linear and one flanking arene from each terphenyl makes a low hapticity contact (η^1 to η^3) with the metal (Fig. 15a), although for Zn(II) and high-spin Mn(II) these contacts are at relatively long distances; (2) when R is Me and R' is Me or H, the MX_2 core is bent and one flanking arene from each terphenyl group interacts with the metal in an η^2 or η^3 mode, resulting in a pseudo-tetrahedral coordination (Fig. 15b), and (3) when R is *i*-Pr and R' is H, the MX_2 core may be either linear or bent, in which case one of the flanking arene groups coordinates in an η^6 mode (Fig. 15c). Although it seems clear that the substituents have some influence on the choice of one of the three alternative structures, one should not rule out the possible existence of stereoisomers.

8. Conclusions

We chemists probably feel more comfortable with 2D representations of the atomic connectivities in molecules than with their spatial arrangement. In part because more than one million crystal structures determined so far, including small molecules, proteins and extended solids, represent only a small fraction of the total number of

chemical compounds known. For that reason, many interesting molecular shapes go unnoticed except for the small group of specialists in a particular area of chemistry. Of course, there are exceptions, and from time to time some innovative or beautiful molecules become widely known, as was the case for buckminsterfullerene, and for the widely developed families of supramolecular knots, rings and cages [45]. Many of the less famous molecules commented in this article have been detected only after hours of careful observation of the molecular structures contained in structural databases such as the Cambridge Structural Database (CSD) and the Inorganic Crystal Structure Database (ICSD). In some cases, the creators of new molecules themselves have called our attention in their communications in a particular way in which we can see them, which helps our visual memory to remind them for the rest of our life.

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