

Calculated »London« π -Electron Magnetic-Properties of Some Carbon-Atom Fragments Extant on the Surface of Icosahedral C_{60} (Buckminsterfullerene)

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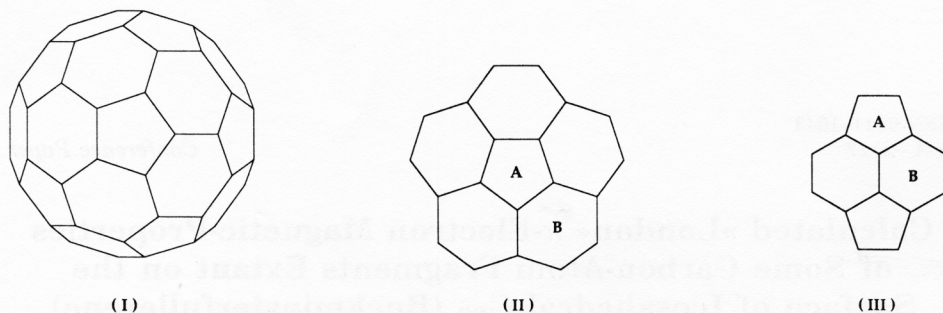
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Semi-empirical calculations, of varying degrees of sophistication, are effected in order to predict the »London« π -electron magnetic-properties of several carbon-atom fragments – some large, some small – that are extant on the spherical surface of the truncated-icosahedral C_{60} cluster known as Buckminsterfullerene. Calculations by the least-approximate of the methods that have been applied predict every one of the Buckminsterfullerene fragments studied to display »London« π -electron magnetic-properties that are, overall, diamagnetic – to the extent of between *ca.* 1/3 of, and *ca.* 5 times, the benzene value.

INTRODUCTION

Since the initial diagnosis of icosahedral C_{60} (Buckminsterfullerene¹ (**I**)) in 1985² – and especially after its subsequent synthesis, in macroscopic quantities, five years later^{3–6} – the magnetic properties of this remarkable species have been the subject of considerable interest, both theoretical^{7–13} (recently spawning some lively controversy in the pages of *Chemical Physics Letters*^{9–12}) and, eventually, experimental.^{5,14,15} In speculating on the structure of other fullerenes (C_n , with n being certain integers), Kroto^{16,17} has observed that an arrangement in which a pentagon is completely surrounded by hexagons is likely to be particularly stable. The same author has further emphasized¹⁶ that corannulene (**II**), a non-planar, saucer-shaped molecule ($C_{20} H_{10}$),



is the prototype system of this kind. Because the carbon-atom skeleton of corannulene (**II**) (like that of pyracylene (**III**)⁸) is formally to be found on appropriate parts of the surface of the Buckminsterfullerene sphere, it is possible that its calculated magnetic-properties might be material to those of the C_{60} cluster.¹⁸

Accordingly, one of the present authors, in the proceedings of an earlier Dubrovnik MATH/CHEM/COMP Conference,¹⁸ reported very rudimentary calculations of the π -electron magnetic-properties of corannulene (**II**). In the present paper, we report some more-sophisticated calculations of these aspects of **II** and we examine also the »London« (»ring-current«) magnetic-susceptibilities of some larger (**IV** and **V**) and smaller (**VI** and **VII**) fragments that are extant on the surface of icosahedral C_{60} (**I**).

METHODS AND CALCULATION

As in our earlier work¹⁹⁻²³ on this subject, six different semi-empirical methods of calculation (labelled 1 to 6) were applied; they are numbered and described in detail in Ref. 21 and are briefly summarized as follows:

Method 1. The London-McWeeny method,²⁴⁻²⁷ based on a simple (»topological«^{19,26}) Hückel²⁸ molecular-orbital (HMO), with resonance integrals fixed at the standard (benzene) value.

Method 2. The London-McWeeny method,²⁴⁻²⁷ based on an iterative (» $\beta\omega'$ »²⁶) HMO²⁹⁻³¹, in which Coulomb integrals are (simultaneously) made iteratively self-consistent with the corresponding calculated bond-orders.

Method 3. The coupled Hartree-Fock, Hall-Hardisson method,³² based on a Pariser-Parr-Pople self-consistent-field (PPP-SCF) wave-function with *fixed* resonance-integrals.

Method 4. The coupled Hartree-Fock Hall-Hardisson method,³² based on a PPP-SCF wave-function (as in Method 3), but this time with *variable* resonance-integrals, iterated so as to be self-consistent with respect to corresponding calculated bond-orders.

Method 5. The method that two of us originally proposed with Coulson;³³ this approach is based on a PPP-SCF wave-function that has *fixed* resonance-integrals.

Method 6. The method of Coulson *et al.*,³³ once again based on a PPP-SCF wave-function (as in Method 5), but now with the difference that the resonance integrals are *variable* and are iterated so as to be self-consistent with respect to the corresponding calculated bond-orders.

As was the case in the discussion of our earlier work,²¹⁻²³ it will be advantageous to bear in mind that methods 1 \rightarrow 3 \rightarrow 5 (in that order) represent increasing sophistication of method (even though these three methods are all based on wave functions that

have *fixed* resonance-integrals). Method 2 → 4 → 6 (also in that order) likewise represent increasing sophistication of method, but these three approaches have the distinct advantage that they are founded on wave functions in which resonance integrals are iterated so as to be self-consistent with respect to the corresponding calculated bond-orders – an especially important feature^{19–23,34,35} when potentially paramagnetic π -electron systems are being dealt with, as is now generally agreed.^{19–23,34,35}

Because Method 6 is the most sophisticated one that we shall be using – and because it will, as a result, give rise to our most reliable predictions of magnetic properties – we draw attention here to the fact that a careful description of Method 6, and of the parametrisation and wave-function on which it is based, has been fully documented in Ref. 21, to which we refer any interested reader requiring more-complete details.

GENERAL ASSUMPTIONS ON WHICH THE CALCULATIONS ARE BASED

The *geometrical* parts of all the above calculations have been based on a model in which all carbon-carbon bond-lengths are assigned the standard benzene-value (1.39 Å), and the areas of five- and six-membered rings are taken to be in the ratio³⁶ 5 cot ($\pi/5$) : 6 cot ($\pi/6$) (*i.e.*, *ca.* 0.6622 : 1.0000). Furthermore, an assumption which, at first sight, might seem a somewhat drastic and unreasonable one has been made: namely that, for the purposes of the magnetic calculations, the species **II**, **IV** and **VI** may be considered to be *planar* (in the geometrical* sense). Now, manifestly, these networks of carbon atoms are *not* geometrically planar for – at least in the context of Buckminsterfullerene, considered here – they lie on the surface of a sphere. However, it has been shown^{39,27} that »ring-current« ideas hold with only minor modification¹⁸ when non-planarity is such that the skeletal distortion around any one bond is comparatively mild. As has been pointed out, because of the way in which strains are spread over many degrees of freedom, this condition does appear to be satisfied in the case of **II**^{40–43} and hydrocarbons like the helicenes,^{44,45} even though the overall non-planarity between well-separated parts of such molecules may in fact be very large.^{39,27,46}

RESULTS AND DISCUSSION

Table I shows the relative π -electron »ring-current« intensities^{24–27} (Method 1), »integrated π -electron current-densities«³³ (Methods 5 and 6) and »London« contributions to magnetic susceptibility^{19–23} (relative to those of the benzene molecule) of the (overall, apparently diamagnetic) π -electron species corannulene (**II**), computed by most of the methods outlined in the previous section. Table II displays, for purposes of comparison, similar calculations (taken from Refs. 21 and 22) relating to the less-diamagnetic pyracylene (**III**). Table III illustrates the corresponding π -electron magnetic-properties of the other Buckminsterfullerene fragments (**IV**, **VI** and **VII**) that we have considered in this study: for the latter, only calculations by the crudest (Method 1) and the most sophisticated (Method 6) of the six semi-empirical approaches we have adopted here are presented in Table III, for reasons of space.

* It may be noted that, in a strictly *graph-theoretical* sense, they are planar – see Refs. 37, 38 for a full discussion of *this* aspect.

TABLE I

Relative π -Electron »Ring-Current« Intensities^{a,24-27,36} (Method 1), »Integrated π -Electron Current-Densities«^{a,33} (Methods 5 and 6) and »London« Contributions^{a,21} to the Magnetic Susceptibility of Corannulene (II) by Various Methods^b

Method of Calculation ^c	»Ring-Current« Intensity (Method 1) »Integrated π -Electron Current-Density« (Methods 5 and 6) (all Relative to Benzene) in Rings		$\chi_{\perp}^{\pi\text{-London}}$ (Corannulene)
	A	B	$\chi_{\perp}^{\pi\text{-London}}$ (Benzene)
1 ^d	0.22	1.22	6.27
3	- ^e	- ^e	5.57
5	-0.01	1.13	5.63
f-----			
4	- ^e	- ^e	4.88
6	-0.04	0.98	5.00

^a Expressed as a *ratio* to the corresponding quantity calculated, by the same Method, for benzene; all calculated values displayed in this Table are, therefore, *dimensionless*. Positive entries indicate diamagnetism and negative entries denote paramagnetism.

^b »Methods 1, 3, 4, 5 and 6 of »Methods of Calculation«.

^c Numbers refer to the labelling of the Method used, according to the scheme described in the section entitled »Methods of Calculation«, where brief details may be found; full details of the assumptions on which the computation in question is based are given in Ref. 21.

^d Results by Method 1 are taken from Ref. 18.

^e Quantity not defined in the context of Methods 3 and 4.

^f The dotted line between the upper and lower sections of the Table divides the results from Methods 1, 3 and 5 – which are based on wave functions that are *not* iteratively self-consistent with respect to resonance integrals and calculated bond-orders – from the results of Methods 4 and 6 which *are* based on wave functions that incorporate such »self-consistency«.

The several calculations on corannulene (II) all support the qualitative conclusion of those based on the crudest of our six approaches (Method 1) that were reported earlier¹⁸ – namely, that this species appears to possess a diamagnetic »London« π -electron magnetic susceptibility ($\chi_{\perp}^{\pi\text{-London}}$ (Species)) which, in size, is of a similar order of magnitude to that of the analogous, unstrained, condensed, benzenoid hydrocarbon with the same number of rings – coronene (Molecule IV of Ref. 18). Method 1 predicts this to be just over six times the benzene value, whilst our least-approximate calculation (*via* Method 6) estimates it to be about five times the benzene value. Methods 3, 4 and 5 – of intermediate sophistication – yield values of $\chi_{\perp}^{\pi\text{-London}}$ (Species) for corannulene (II) of between *ca.* 4.8 and 5.7 times the corresponding quantity for benzene. As for the individual »ring-current« intensities (Method 1) and »integrated π -electron current-densities« (Methods 5 and 6) in each of the rings **A** and **B** of corannulene (II), the magnitude of the latter in the five-membered ring (**A**) is (according to Method 6, our best method) virtually zero, while those in the six-membered rings (**B**) are very nearly equal to the benzene value. These calculations by Methods 1, 3 and 5 (non-iterative) and by Methods 4 and 6 (iterative) also well illustrate the by-now very familiar conclusion¹⁹⁻²³ that the predicted »London« π -electron magnetic-properties of those species which Method 1 diagnoses as unambiguously diamagnetic in this regard (especially if such diamagnetic properties are of the same order of magnitude as those of the neutral, condensed, benzenoid hydrocarbons of similar size) are remarkably *insensitive* to whether an iterative or a non-iterative method has been used to calculate them.

By contrast, Table II reminds us that predictions about the »London« π -electron magnetic properties of pyracylene (III) are *very* sensitive to the method used in making

TABLE II

Relative π -Electron »Ring-Current« Intensities^{a,24-27,36} (Methods 1 and 2), »Integrated π -Electron Current-Densities«^{a,21} to Magnetic Susceptibility Perpendicular to the Molecular Plane of Pycrylene (**III**), by Various Methods^b

Method of Calculation ^c	»Ring-Current« Intensity (Methods 1 and 2), »Integrated π -Electron Current-Density« (Methods 5 and 6) (all Relative to Benzene) in Rings		$\chi_{\perp}^{\pi\text{-London}}$ (Pycrylene)
	A	B	$\chi_{\perp}^{\pi\text{-London}}$ (Benzene)
1 ^d	-2.32	-0.39	-3.84
3 ^d	- ^e	- ^e	-3.25
5 ^d	-2.16	-0.20	-2.93
f	-----	-----	-----
2 ^d	-1.01	0.31	-0.73
4 ^d	- ^e	- ^e	0.15
6 ^d	-0.74	0.56	0.31

^a Expressed as a *ratio* to the corresponding quantity calculated, by the same Method, for benzene; all calculated values displayed in this Table are, therefore, *dimensionless*. Positive entries indicate diamagnetism and negative entries signify paramagnetism.

^b Methods 1 - 6 of »Methods of Calculation«.

^c Numbers refer to the labelling of the Method used, according to the scheme described in the section entitled »Methods of Calculation«, where brief details may be found; full details of the assumptions on which the computation in question is based are given in Ref. 21.

^d Results taken from Refs. 21 and 22.

^e Calculation not available; (quantity not defined in the context of Methods 3 and 4).

^f The dotted line between the upper and lower sections of the Table divides the results from Methods 1, 3 and 5 - which are based on wave functions that are *not* iteratively self-consistent with respect to resonance integrals and calculated bond-orders - from the results of Methods 2, 4 and 6, which *are* based on wave functions that incorporate such »self-consistency«.

them. This same remark applies to other systems which, on the basis of a simple, non-iterative Hückel-McWeeny calculation (Method 1), are expected to be predominantly paramagnetic in their computed »ring-current« properties.^{19-23,34,35} In particular (as we have emphasized on several occasions previously¹⁹⁻²³), it matters very much whether or not the wave function on which the calculations are based takes variation of resonance integrals into account. We have, therefore, earlier stressed quite earnestly¹⁹⁻²³ that resonance-integral variation with respect to calculated³⁴ (or experimental^{21,35}) bond-length is *vital* in order to avoid spuriously exaggerated²² predictions of the extent of paramagnetism in systems of this type. There is now general agreement on this matter.^{19-23,34,35,47-50} Accordingly, the results by Methods 2, 4 and 6 are probably the only ones that should be taken seriously, among those presented in Table II, and we do in fact further confine our subsequent discussion to the results from our best method, Method 6. It is seen that, on the basis of the calculations effected by this latter approach, the six-membered ring (**B**) is diamagnetic, to the extent of just over half the benzene value, while - even according to this iterative calculation which, furthermore, has made minimal appeal to the London integral-approximation^{33,27} - the five-membered ring (**A**) has finally settled to be paramagnetic, to the extent of *ca.* 3/4 of the magnitude of the diamagnetic »integrated π -electron current-density« calculated, by the same method, for benzene. Even so, the overall »London« π -electron magnetic-susceptibility of pycrylene (**III**) is predicted to be (just) diamagnetic - at a little under 1/3 of the benzene value.

When choosing some larger fragments that form part of the surface of the Buckminsterfullerene sphere it would appear to be of interest to consider a species such as **IV**, which is approximately the extent of the complete icosahedral C_{60} structure that can be seen when a model of it is observed by the human eye. The fragment shown as **IV** is the view that would be seen when the sphere presents itself with a pentagonal face (**A**) at the centre. In this particular orientation (**IV**), corannulene (**II**) is the most important feature because (a) the faces which form the outer perimeter are at an angle of more than 60° to face **A**, and (b) the corannulene unit is a particularly stable one. (It may be observed that fragment **IV** represents precisely half the Buckminsterfullerene structure: it comprises six of the twelve pentagons, and ten of the twenty hexagons; it also contains twenty interior carbon-atoms and twenty on the »perimeter« – which latter are, of course, »shared« with the species corresponding to **IV** extant on the *other*, more-distant, side of the sphere).

The first two lines of Table III show our calculations on **IV** by Method 1 (our crudest method) and Method 6 (our best method). As can be seen there, the »London« π -electron magnetic-susceptibility of this species is predicted to be strongly paramagnetic by the non-iterative Method 1, and diamagnetic (to the extent of about a 30% increase on the benzene value) by the more-realistic iterative calculation represented by Method 6. All rings are predicted to be paramagnetic by the non-iterative calculations (Method 1) but, on iteration (Method 6), the six-membered rings (**B** and **D**), as expected, become diamagnetic (to the extent of about half the benzene value), while the five-membered rings stay (just) paramagnetic. Overall, though, the »London« π -

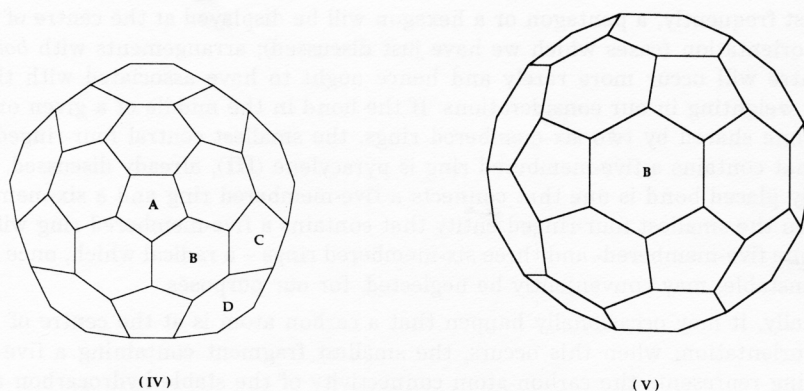
TABLE III

*Relative π -Electron »Ring-Current« Intensities^{a,24-27,36} (Method 1), Integrated π -Electron Current-Densities^{a,33} (Method 6), and »London« Contribution^{a,21} to Magnetic Susceptibility in (a) Species **IV**, (b) Species **VI**, and (c) Acenaphthylene **VII***

Method of Calculation ^b	»Ring-Current« Intensity (Method 1), »Integrated π -Electron Current-Density« (Method 6) (all Relative to Benzene) in Rings				$\chi_{\perp}^{\pi\text{-London}}$ (Species)
	A	B	C	D	$\chi_{\perp}^{\pi\text{-London}}$ (Benzene)
(a) Species (IV)					
1	-1.14	-0.33	-1.98	-0.33	-10.60
6	-0.17	0.52	-0.49	0.44	1.30
(b) Species (VI)					
1	-1.19	-0.29	0.79	0.24	-0.61
6	-0.43	-0.11	0.68	0.82	1.22
(c) Acenaphthylene (VII)					
1	0.10	0.93	-	-	2.54
6	1.31	1.32	-	-	3.55

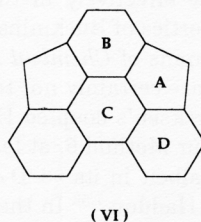
^a Expressed as a *ratio* to the corresponding quantity calculated, by the same Method, for benzene; all calculated values displayed in this Table are, accordingly, *dimensionless*. Positive entries denote diamagnetism and negative entries signify paramagnetism.

^b Numbers refer to the labelling of the Method used, according to the scheme described in the section entitled »Methods of Calculation«, where brief details may be found; full details of the assumptions on which the computation in question is based are given in Ref. 21.



electron contribution to the magnetic susceptibility of **IV** is predicted to be diamagnetic, and, in size, about 1.3 times the benzene value.

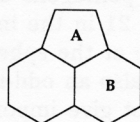
Now it would appear that an entity analogous to **IV** but with a hexagonal face (**B**) at the centre ought also to be considered (and, in fact, since the ratio of hexagons to pentagons in **I** is 20 : 12, it might be argued that this orientation ought to be given a weighting of 5/3 times that of **IV**. However, such a species **V**, in isolation, is a radical which, though again comprising six pentagons and ten hexagons, possesses an odd number (39) of carbon atoms: it has 21 in the interior and 18 (shared with the corresponding species on the other side of the sphere) around the periphery. The first corona around the central ring **B** is also an odd-membered entity and we feel, therefore, that it, also, should likewise not give important contributions to the magnetic quantities we are seeking to calculate, because of its consequent instability. It becomes stable, however, if one of the pentagons is opened by suppressing a single carbon atom: we might, therefore, usefully consider a species such as **VI**, thus obtained.



The results of our computations on **VI** are presented in the middle two lines of Table III. Although **VI** is predicted to have a marginally paramagnetic »London« π -electron magnetic-susceptibility on the basis of the non-iterative calculation by Method 1, this susceptibility settles, on iteration (Method 6), to a »normal« diamagnetic value of *ca.* 1.2 times the corresponding quantity calculated, by the same method, for benzene. The »integrated π -electron current-density« calculated by Method 6 for the five-membered ring (**A**) remains paramagnetic (-0.43), as does (marginally) that for *one* of the six-membered rings (**B**), the other two six-membered rings (**C** and **D**) remaining diamagnetic at *ca.* 68% and 82%, respectively, of the benzene value.

Most frequently, a pentagon or a hexagon will be displayed at the centre of a particular orientation (cases which we have just discussed); arrangements with *bonds* at the centre will occur more rarely and hence ought to have associated with them a smaller weighting in our considerations. If the bond in the middle of a given orientation is one shared by two six-membered rings, the smallest central four-ringed fragment that contains a five-membered ring is pyracylene (**III**), already discussed. If the centrally placed bond is one that connects a five-membered ring and a six-membered ring then the smallest four-ringed entity that contains a five-membered ring will consist of one five-membered- and three six-membered rings – a radical which, once again, being unstable, may conveniently be neglected, for our purposes.

Finally, it may occasionally happen that a carbon atom is at the centre of a particular orientation; when this occurs, the smallest fragment containing a five-membered ring represents the carbon-atom connectivity of the stable hydrocarbon acenaphthylene (**VII**). Calculations by Methods 1 and 6 (presented in the last two lines of Table III) show this to be qualitatively diamagnetic, with »ring currents« and »integrated π -electron current-densities« both being diamagnetic (about 30% more than the benzene value, according to the more-reliable Method 6). The »London« contribution to its π -electron magnetic susceptibility, perpendicular to its molecular plane, is *ca.* 2.5 – 3.5 times this quantity calculated, by the same method, for benzene.



(VII)

We do not feel that our semi-empirical computations are of such a quality as to enable us to presume to adjudicate effectively in the entertaining and lively debate about the calculated magnetic-properties of Buckminsterfullerene (**I**) which has recently been carried on through the columns of *Chemical Physics Letters*.⁹⁻¹² The semi-empirical methods that we have used are certainly not to be compared with the »iterative AO version« of McWeeny and Diercksen's coupled Hartree-Fock formalism⁵¹ used by Fowler *et al.* in Ref. 9 – although our Method 6, at least, is probably of greater sophistication than the HMO approach (albeit in its »3-D« form^{52-56,7}) originally applied to Buckminsterfullerene by Elser and Haddon.^{7,56} In these circumstances, we merely conclude by noting that calculations by the least-approximate of those methods that we have used (what we have referred to throughout as »Method 6«) predict the »London« π -electron magnetic-susceptibilities of every one of the Buckminsterfullerene fragments considered here to be, overall, diamagnetic – ranging from *ca.* 1/3 of the benzene value in the case of pyracylene (**III**) to *ca.* 5 times the benzene value in the case of corannulene (**II**), the species **IV**, **VI** and **VII** being intermediate between these two extremes.

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SAŽETAK

**Račun »Londonovih« π -elektronskih magnetskih svojstava
nekih ugljičnih fragmenata prisutnih u ikozaedarskom
C₆₀ (buckminsterfullerenu)**

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Semi-empirijski postupci, različitog stupnja složenosti, upotrijebljeni su za proračun »Londonovih« π -elektronskih magnetskih svojstava ugljičnih fragmenata prisutnih u molekuli buckminsterfullerena. Najtočnijim od primijenjenih postupaka predviđa se da će svaki od ispitivanih fragmenata imati dijamagnetska svojstva, u iznosu koji se kreće od 1/3 do peterostruke vrijednosti za benzen.