Combinatorics of Edge Symmetry: Chiral and Achiral Edge Colorings of Icosahedral Giant Fullerenes: $C_{80}$, $C_{180}$, and $C_{240}$ †

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† Dedicated to the memory of Prof. Jun-Ichi Aihara, a pioneer in superaromaticity and aromaticity.

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Abstract: We develop the combinatorics of edge symmetry and edge colorings under the action of the edge group for icosahedral giant fullerenes from $C_{80}$ to $C_{240}$. We use computational symmetry techniques that employ Sheehan’s modification of Pólya’s theorem and the Möbius inversion method together with generalized character cycle indices. These techniques are applied to generate edge group symmetry comprised of induced edge permutations and thus colorings of giant fullerenes under the edge symmetry action for all irreducible representations. We primarily consider high-symmetry icosahedral fullerenes such as $C_{80}$ with a chamfered dodecahedron structure, icosahedral $C_{180}$, and $C_{240}$ with a chamfered truncated icosahedron geometry. These symmetry-based combinatorial techniques enumerate both achiral and chiral edge colorings of such giant fullerenes with or without constraints. Our computed results show that there are several equivalence classes of edge colorings for giant fullerenes, most of which are chiral. The techniques can be applied to superaromaticity, sextet polynomials, the rapid computation of conjugated circuits and resonance energies, chirality measures, etc., through the enumeration of equivalence classes of edge colorings.

Keywords: edge symmetry of giant fullerenes; edge colorings of giant icosahedral fullerenes; chirality in edge colorings of giant fullerenes; combinatorial techniques for giant fullerenes; aromaticity and superaromaticity

1. Introduction

Symmetry, combinatorial enumerations, chirality, symmetry-based reactivity, spectroscopy, and the synthesis of fullerenes have been a subject of study over the years [1–42]. Whereas the vertex groups or the automorphism groups of graphs of fullerenes, point group symmetries of molecules, solids, and fullerenes have been quite well studied [1–4,20,24,26,29–39], this is not the case with the edge symmetry or edge groups, especially concerning giant fullerenes. Although the two symmetry groups are isomorphic to each other, the induced permutations on the edges of the associated molecular graphs by vertex permutations are not always obvious or easily obtained. Such an edge group comprising of edge permutations that preserve the connectivity of the fullerene graph is required for many applications, including edge colorings under the group action. Then, special cases of equivalence classes of edge colorings correspond to equivalence classes of conjugated circuits or resonance structures [43–53] and thus can provide an efficient platform for the rapid computation of resonance energies, conjugated circuits, and other properties that provide insight into aromaticity and superaromaticity [43–53]. Fullerenes are a special class of compounds containing only carbons with 12 pentagons and a varied number of hexagons. Fullerenes are especially interesting candidates for the edge colorings, as they contain a network of alternating arrangements of single and double bonds, which results in a large number of resonance structures or conjugated circuits [43–53], especially for giant
fullerenes. Although one can employ high-level ab initio quantum chemical computations on giant fullerenes with supercomputers, techniques such as conjugated circuit theory and the combinatorial enumeration of resonance structures provide a viable and efficient alternating platform for shedding light into the structures and reactivity [43–53] of polycyclic aromatics and thus potentially giant fullerenes. Such combinatorial enumerations have been of interest [54–59], and especially Kekulé addition patterns related to edge colorings of smaller fullerenes [60].

Combinatorial enumeration of Kekulé structures, conjugated circuits, and resonance structures of both acyclic and cyclic polyacenes have been the topic of several studies [43–53], as such techniques aid in the computation of topological resonance energies, stabilities, and reactivities of polycyclic, acyclic aromatics, and fullerenes. We note that the various Kekulé structures are special cases of edge colorings of the associated graphs, and thus the enumeration of equivalence classes of edge colorings aid in the simplification of such computations, as all members of a given equivalence class would make the same contribution to the topological resonance energies or the stabilities of fullerenes or polycyclic aromatic compounds.

Experimental studies on fullerenes and giant fullerenes have been focused on the synthesis, structures, chirality, reactivity, and spectroscopy of fullerenes and their derivatives [5–28]. Giant fullerene cages and onions of fullerenes are interesting because they can be employed to encapsulate metallic oxides as well as small clusters inside fullerenes such as $C_{180}$ and $C_{240}$ [5–7,20–23,28], and such cages find a variety of applications including the environmental remediation of high-level nuclear wastes [61–63]. The chirality of substituted fullerenes such as chlorinated fullerenes and their spectroscopy has also received attention [4,21,24,26,30,33]. The edge connectivity of fullerenes can shed light not only on the halogenation reactions but also on chemisorption. The existence of inequivalent edges in fullerenes would provide an opportunity to study reactivity contrast upon the addition of molecules such as $H_2$, $N_2$, and $O_2$, etc., as inequivalent edges can result in different reactivity patterns. Thus, edge colorings under the edge group action of giant fullerenes can provide insights into reactivity patterns especially in chemisorptive reactions and reactions that involve the attachment of molecules to the edges of fullerenes. The icosahedral giant series of fullerenes with the formula $C_{60k}$ for $k = m^2$ of which $C_{60}$, $C_{240}$, $C_{340}$, $C_{360}$, etc., are members are especially interesting for their high symmetry and chirality of derivatives of these members. The enumeration of the isomers of polysubstituted giant icosahedral fullerenes was considered by the present author in a recent study [4]. Subsequently, stimulated by the pentagonal face dynamics of nanocones and chirality concepts, Balsubramanian et al. [64] recently outlined a combinatorial scheme for the face colorings of icosahedral giant fullerenes. The present study complements the previous two related works [4,64] in that this is the first study on the edge colorings of giant fullerenes, which is a topic of considerable interest in the context of reactivity, stability, enumeration of Kekulé structures, conjugated structures, achirality, and chirality arising from the edge colorings and edge groups etc., of giant fullerenes. The edge colorings considered here are also relevant to heterofullerenes such as $C_{38}N_{12}$ [37,65].

Stimulated by the intriguing symmetry, chirality, edge substitution, and reactivity of giant fullerenes, in the present study, we undertake edge symmetry and edge colorings under the group action for a series of giant fullerenes, namely $C_{30}$, $C_{180}$, and $C_{240}$. We show that the Möbius inversion technique combined with our generalization of Sheehan’s modification of Pólya’s theorem to all characters yields powerful combinatorial generating functions for the edge colorings of giant fullerenes for all of the irreducible representations of the symmetry group of fullerenes. We focus on the edge colorings of high symmetry icosahedral giant fullerenes that have been of both experimental and theoretical interest.

## 2. Möbius Inversion Technique with Generalization of Sheehan’s Theorem to All Characters as Tools for the Combinatorics of Edge Groups and Edge Colorings of Giant Fullerenes

The key advantage of the Möbius inversion technique in the context of cycle index polynomials is that when a group acts on two different sets and if the cycle types for the permutations for the action
on the smaller sets are known, then the technique can be used to obtain the cycle types of a larger set of objects. The application of the technique results in the inversion of a polynomial generating function from one set to the other through the divisors, which we used extensively in the context of hypercube enumerations for coloring the various hyperplanes for all of the irreducible representations of the hyperoctahedral group \([55]\). That is, the coefficient of \(x^q\) in the inverted polynomial generating function \(Q_p(x)\) shown below obtained from a known polynomial \(F_d(x)\) generates the permutational cycle types for a larger set from a smaller set—for example, the edges of a 7D hypercube from the permutational matrix types for the hyperoctahedral group \([55]\).

\[
Q_p = \sum_{d|p} \mu(p/d) F_d(x),
\]  

(1)

where the sum is over all divisors \(d\) of \(p\), and \(\mu(p/d)\) is the Möbius function that yields the cycle types of larger sets, where \(F_d(x)\) is the known polynomial in \(x\) as constructed from the permutations of objects of a smaller set with the Group \(G\) commonly acting on both sets. The Möbius function, which in the above equation is denoted as \(\mu(p/d)\), takes values 1, \(-1\), \(-1\), 0, \(-1\), 1, \(-1\), 0, 0, 1 \ldots for arguments 1 to 10. The Möbius function for any number is 0, +1, or \(-1\) depending on whether it is a perfect square and the number of prime factors in the argument.

Figure 1 shows a regular icosahedron, a vertex–edge combo coloring of the icosahedron with 5 different colors, the \(C_{20}\) dodecahedron, and the \(C_{60}\) buckminsterfullerene—all of which exhibit the icosahedral symmetry. Fullerenes are vertex regular graphs with 3 vertex degrees that contain 12 pentagons. Therefore, for any fullerene, the number of edges is given by \(3n/2\), where \(n\) is the number of vertices. For example, there are 360 edges in the case of the \(C_{240}\) icosahedral fullerene. Each of the proper and improper rotational operations of the icosahedron induces a permutation on the set of 360 edges of \(C_{240}\), a permutation on the 240 vertices, and a permutation on 122 faces of the giant \(C_{240}\) fullerene with the icosahedral point group symmetry. Euler’s formula connects the number of vertices, edges, and faces of the fullerene; thus, we can obtain the number of any one of them when the other two are known. The group action on the edges of the fullerene can be obtained through a number of techniques, including the generation of the edge automorphism group from the vertex automorphism group of the graph. However, we note that the permutational cycle types of group action on the edges can be obtained computationally through the Möbius sum. It can also been seen that certain operations such as the \(C_5\) rotational axis for an icosahedral fullerene cage pass through the center of a pentagon (see Figure 2e) and not through any of the vertices with the exception that for the 12-vertex icosahedron (see Figure 1a,b), the \(C_5\) axis passes through two opposite vertices of the icosahedron. Hence, this is directly related to the Möbius concept of whether the fold-ness of the rotational axis divides the number of objects in the set or not. For example, for the \(C_5\) axis, the number 5 does not divide 12 in the case of the vertices of an icosahedron, thus leaving a remainder of 2 when 5 is divided by 12. This necessitates the \(C_5\) axis to pass through two vertices that are across each other for the icosahedron. For giant fullerenes such as \(C_{240}\), the \(C_5\) axis passes through the center of a pentagon, and as 5 is a divisor of 240, it is clear that the vertex permutation will be described by \(5^{48}\). As seen from Table 1, the conjugacy class representatives of the icosahedral fullerene group comprises the set \(\{E, C_6, C_5, C_3, C_2, C_1, S_{10}, S_{12}, S_6, \sigma_d\}\). For all of the icosahedral fullerenes considered here, all the edges are not equivalent or alternatively, the edges are divided into more than one equivalence class for such giant fullerenes. In such a scenario, it is more suitable to consider a generalization of Pólya’s theorem outlined by Sheehan \([54]\), which we further generalize to all characters and the edge action of the icosahedral group.
Figure 1. A regular icosahedron (a), a combo of vertex–edge coloring of the icosahedron with 5 colors (b), where an edge incident on two vertices is defined with the corresponding vertex colors—for example, blue-red edge (b), C\textsubscript{20} dodecahedral fullerene (c), and C\textsubscript{60} Buckminsterfullerene (d)—all with Ih icosahedral symmetry. The C\textsubscript{5} axes pass through each vertex of the icosahedron colored to show the pentagon, and the C\textsubscript{3} axes pass through the center of triangular face, while the C\textsubscript{2} axes pass through the center of an edge.
(b) Icosahedral $C_{180}$

(c) Icosahedral $C_{240}$

(d) Two views of $C_{240}$; the right one shows the $C_5$ axis passing through the center of a pentagon, while the $C_3$ axis shown on the right panel passes through the center of a cyan hexagon.

(e) $C_{240}$ showing pentagons in red, two types of hexagons in green and yellow.

Figure 2. Structural representations for $C_{80}$, $C_{180}$, and $C_{240}$ fullerene cages considered here for edge colorings. Panel (d) is reproduced from [https://robertlovespi.net/](https://robertlovespi.net/) and it is copyright-free.
Table 1. Character table of 1h, permutation cycles and their lengths for the 60 pent–hex edges and remaining hex–hex edges of C_{60} (1h), Icosahedron, C_{80} (1h), C_{180} (1h), and C_{240} (1h) upon action on edges.a

| CC                        | E       | 15C_2 | 12C_3 | 12C_3^2 | 20C_3 | I       | 12S_{10} | 12S_{10}^3 | 20S_4 | 15S_4 |
|---------------------------|---------|-------|-------|---------|-------|---------|----------|------------|--------|--------|
| C_{60} (pent–hex)         | 1^{10}  | 2^{14} | 5^{1} | 5^{1}   | 3^{10} | 2^{15}  | 10^{3}   | 10^{3}     | 6^{5}  | 1^{2}  |
| C_{60} (pent–hex; edges)  | 1^{10}  | 2^{10} | 5^{12} | 5^{12}  | 3^{10} | 2^{15}  | 10^{6}   | 10^{6}     | 6^{10} | 1^{4}  |
| C_{180} (pent–hex; edges) | 1^{10}  | 2^{10} | 5^{12} | 5^{12}  | 3^{10} | 2^{15}  | 10^{6}   | 10^{6}     | 6^{10} | 1^{4}  |
| C_{240} (pent–hex; chamfered dodecahedron) | 1^{10}  | 2^{10} | 5^{12} | 5^{12}  | 3^{10} | 2^{15}  | 10^{6}   | 10^{6}     | 6^{10} | 1^{4}  |
| A_4                        | 1       | 1     | 1     | 1       | 1     | 1       | 1        | 1          | 1      | 1      |
| T_{1g}                     | 3       | −1    | ω     | ω^*    | 0     | 3       | ω        | 0          | −1     | −1     |
| T_{2g}                     | 3       | −1    | ω^*   | ω      | 0     | 3       | ω        | ω^*        | 0      | −1     |
| G_2                        | 4       | 0     | −1    | −1     | −1    | 4       | −1       | −1         | 1      | 1      |
| H_3                        | 5       | 1     | 0     | 0       | −1    | 5       | 0        | 0          | −1     | 1      |
| A_6                        | 1       | 1     | 1     | 1       | 1     | 1       | −1       | −1         | −1     | −1     |
| T_{1u}                     | 3       | −1    | ω     | ω^*    | 0     | −3      | −ω       | −ω^*       | 0      | 1      |
| T_{2u}                     | 3       | −1    | ω^*   | ω      | 0     | −3      | −ω       | −ω^*       | 0      | 1      |
| G_8                        | 4       | 0     | −1    | −1     | −1    | −4      | 1        | 1          | 1      | 0      |
| H_4                        | 5       | 1     | 0     | 0       | −1    | −5      | 0        | 0          | 1      | −1     |

ω = (1 + √5)/2 (golden ratio). ω = (1 − √5)/2. a Permutation cycles of 30 pent–hex edges of all fullerenes are shown only once, as they are the same for all icosahedral fullerenes with 12 isolated pentagons.

Consider a set D consisting of the edges of a fullerene, and because for all the fullerenes considered here, there exists more than one equivalence classes of edges, we further partition the edge set D of fullerenes into two sets Y_1, Y_2, ..., Y_m where Y_i is an equivalence class partition of the edges of the fullerene. That is, ∪_{i=1}^{m} Y_i = D, Y_i ∩ Y_j = ∅, for i ≠ j, where m = number of the edges of the fullerene cage.

The action of a group G, in this case the icosahedral group of 120 operations, on the set D, induces a permutation in various Y_i sets such that every cycle is contained within the set Y_i. Consequently, an edge coloring of the fullerene can be considered as a map from D to R, where R is the set of colors, but because D is partitioned into various equivalence classes, one may choose an independent set of colors for each set Y_i. Sheehan [54] has considered such a generalization of Pólya’s theorem for the enumeration under group action for the case where the set D is partitioned into multiple sets. We note that Sheehan’s generalization of Pólya’s theorem reduces even the Redfield–Read superposition theorem to a special case. Sheehan’s technique [54] is further generalized here to all characters of the irreducible representations of the icosahedral group for the edge colorings of fullerenes. These techniques also yield the number of chiral and all achiral colorings of the fullerene cages for a different distribution of colors.

The generalized character cycle index (GCCCI) polynomial for any character of the automorphism group G of the fullerene cage under consideration is defined as

\[ P_G^x = \frac{1}{|G|} \sum_{g \in G} \chi(g) \prod_i \prod_j x^{g_{ij}(g)} \]  

(2)
where the sum is over all the permutation representations of $g \in G$, and $c_i(g)$ is the number of $j$-cycles of $g \in G$ upon its action on the set $Y_i$. That is, the second index $j$ in $c_i(g)$ stands for the length of the cycle (that is a $j$-cycle) generated in the $Y_i$ set upon the action of $g \in G$. We also note that unlike the ordinary cycle index of Pólya’s enumeration theorem, the generalization considered here generates a GCCI for each irreducible representation of the $I_h$ group for the icosahedral fullerene. Consequently, we employ an even more powerful generating function than the ordinary Pólya or the Sheehan cycle index [54] to enumerate the edge colorings of giant fullerenes for each irreducible representation (IR).

Define $[n]$ as an ordered partition, or the composition of $n$ into $p$ parts such that $n_1 \geq 0, n_2 \geq 0, \ldots, n_p \geq 0, \sum_{i=1}^{p} n_i = n$. We can obtain a multinomial generating function (GF) in $\lambda$s with $n_1$ colors of the type $\lambda_1, n_2$ colors of the type $\lambda_2 \ldots n_p$ colors of the type $\lambda_p$, which are defined as follows:

$$
(\lambda_1 + \lambda_2 + \cdots + \lambda_p)^n = \sum_{[n]} \binom{n}{n_1 n_2 \ldots n_p} \lambda_1^{n_1} \lambda_2^{n_2} \ldots \lambda_{p-1}^{n_{p-1}} \lambda_p^{n_p} \tag{3}
$$

where $\binom{n}{n_1 n_2 \ldots n_p}$ are multinomials given by

$$
\binom{n}{n_1 n_2 \ldots n_p} = \frac{n!}{n_1! n_2! \ldots n_{p-1}! n_p!}. \tag{4}
$$

Let $R$ be a set of colors partitioned into sets $R_1, R_2 \ldots R_m$ with the same number of partitions as the $Y$ sets such that $R = R_1 \cup R_2 \cup \ldots R_m$ and $R_i \cap R_j = \emptyset$. Furthermore, let $w_{ij}$ be the weight of each color $r_i$ in the set $R_i$. The generating function for each irreducible representation for the edge colorings of giant fullerene cages considered here is given by

$$
GF\left(\lambda_1, \lambda_2 \ldots \lambda_p\right) = P_G^{R}\left[\mathbf{s}_k \rightarrow \left(\mathbf{w}_{r_i}^k + \mathbf{w}_{r_2}^k + \ldots + \mathbf{w}_{r_{p-1}}^k + \mathbf{w}_{r_p}^k\right)\right]. \tag{5}
$$

where $p_i = |R_i|$. The GFs obtained above for each IR and various color distributions provides the number of edge coloring with varying color distribution for the different equivalence classes of the edges, independent of each other and those that transform according to the irreducible representation whose character is given by $\chi$. Thus, all of the techniques demonstrated including the Möbius function method, GCCCs, and generating functions were all programmed into FORTRAN ’95 codes using the quadruple precision arithmetic as the number of edge colorings explode combinatorially. We note that in order to make the codes efficient, especially for larger fullerenes, we have taken considerable efforts to optimize the codes by designing efficient algorithms for the computations of multinomial and binomial numbers using recursion as opposed to an explicit evaluation of factorials and identifying the equivalence of multinomials, thereby eliminating duplicative computations. Furthermore, although we have obtained the results for all of the irreducible representations in the ensuing section, we show the results only for the chiral and totally symmetric irreducible representations, as these are the most interesting cases for the present study. However, we point out that the results obtained for other irreducible representations are quite important in several other applications such as the enumeration of nuclear spin functions, nuclear spin statistics, Electron Spin Resonance (ESR) spin function enumerations for the assignment of the observed hyperfine structures, and so on [57–59]. Thus, there is a wealth of information latent in these generating functions for the various irreducible representations for giant fullerenes.

3. Results and Discussions

Table 1 displays the various conjugacy classes of the icosahedral group, the permutation orbit structures for the edges for $C_{80}$ ($I_h$), $C_{180}$ ($I_h$), and $C_{240}$ ($I_h$), and the character values of the $I_h$ group under each conjugacy class. The three structures considered here are shown in Figure 2 together with
other structures of icosahedral symmetry in Figure 1. Among the fullerenes considered here, we illustrate the procedure of constructing the GCCIs with \( C_{80} (I_h) \) (the chamfered dodecahedron shown in Figure 2), the related edges of the icosahedron in Figure 1a,b, and the corresponding generating functions. There are two types of edges in \( C_{80} \) as seen from Figure 2 analogous to buckminsterfullerene \( C_{60} \) (Figure 1d): the edges that are shared by a pentagon and hexagon are distinct from the edges shared by two hexagons. Thus, for the case of \( C_{80} \), there are two equivalent class edges: namely, \( Y_1 \) and \( Y_2 \). Let the first set correspond to the edges shared by a pentagon and hexagon. As there are exactly 12 pentagons in any fullerene, it is evident that the cardinality of the set \( Y_1 \) is 60. This leaves 60 edges that are common to two hexagons in the case of \( C_{80} \); or, the cardinality of the set \( Y_2 \) is also 60 for the case of \( C_{80} \). However, as can be seen from Table 1, the cardinalities of sets \( Y_1 \) and \( Y_2 \) are 60 and 30, respectively for the buckminsterfullerene case. From Table 1, we combine the cycle types that are shown for the pent–hex edges together with the cycle types for \( C_{80} \) (hex–hex) edges to obtain the GCCIs for the \( A_g \) and \( A_u \) irreducible representations of the \( I_h \) group shown below:

\[
P_{A_g}^{C_{60}} = \frac{1}{120} \{ 1^{60,30} + 15 1^{30,30} \}_{11,21}^{13,22} + 24 s_{11,15,22}^{12,22} + 20 s_{13,23}^{20,20} + s_{12,22}^{30,30} + 24 s_{1,10,2,10}^{6,6} + 20 s_{16,26}^{6,6} \}
\]

\[
P_{A_u}^{C_{60}} = \frac{1}{120} \{ 1^{60,30} + 15 1^{30,30} \}_{11,21}^{12,22} + 24 s_{12,22}^{12,12} + 20 s_{13,23}^{20,20} - s_{12,22}^{30,30} - 24 s_{1,10,2,10}^{6,6} - 20 s_{16,26}^{6,6} - 15 s_{11,12,21,22}^{12,4,28} \}
\]

We note that it is coincidental that the cardinalities of both equivalence classes for the edge set of \( C_{80} \) became the same and equal to 60, which is not the case in general. For the \( C_{60} \) buckminster fullerene, the cardinalities of the two sets are 60 and 30, respectively, and thus, we obtain different terms for \( s_{11} \) and \( s_{21} \) in each of the terms of the GCCIs for \( C_{60} \) for the \( A_g \) and \( A_u \) IRs, as shown below:

\[
P_{A_g}^{C_{60}} (C_{60}) = \frac{1}{120} \{ 1^{60,30} + 15 1^{30,30} \}_{11,21}^{12,22} + 24 s_{12,22}^{12,12} + 20 s_{13,23}^{20,20} + s_{12,22}^{30,30} + 24 s_{1,10,2,10}^{6,6} + 20 s_{16,26}^{6,6} + 15 s_{11,12,21,22}^{12,4,28} \}
\]

\[
P_{A_u}^{C_{60}} (C_{60}) = \frac{1}{120} \{ 1^{60,30} + 15 1^{30,30} \}_{11,21}^{12,22} + 24 s_{12,22}^{12,12} + 20 s_{13,23}^{20,20} - s_{12,22}^{30,30} - 24 s_{1,10,2,10}^{6,6} - 20 s_{16,26}^{6,6} - 15 s_{11,12,21,22}^{12,4,28} + 13 \}
\]

Generating functions for the two cases with 4 different colors to color the edges common to a pentagon and hexagon (denoted as pent–hex edges) and 3 different colors (say red, green, and blue) to color the edges common to two hexagons (denoted as hex–hex) for the \( A_g \) of \( C_{60} \) are shown first in Equation (10).

\[
GF_{A_u}^{C_{60}} = \frac{1}{120} \{ 1 + a + b + c \}^{60} (1 + d + e)^{30} + 15 (1 + a^2 + b^2 + c^2)^{30} (1 + d + e)^{30} (1 + d^2 + e^2)^{14} + 24 (1 + a^5 + b^5 + c^5)^{12} (1 + d^5 + e^5)^6 + 20 (1 + a^3 + b^3 + c^3)^{20} (1 + d^3 + e^3)^{10} \}
\]

\[
+ 24 (1 + a^{10} + b^{10} + c^{10})^6 (1 + d^{10} + e^{10})^3 + 20 (1 + a^6 + b^6 + c^6)^{10} (1 + d^6 + e^6)^3 + 15 (1 + a + b + c)^4 (1 + a^2 + b^2 + c^2)^{28} (1 + d + e)^4 (1 + d^2 + e^2)^{13} \}
\]
The corresponding GF for the $A_u$ IR is obtained in an analogous manner, and it shown as Equation (11):

$$GF_{G}^{A_u} = \frac{1}{120} \left\{ 1 + (a + b + c)^{60}(1 + d + e)^{30} + 15(1 + a^2 + b^2 + c^2)^{30}(1 + d + e)^2(1 + d^2 + e^2)^{14} + 24(1 + a^5 + b^5 + c^5)^{12}(1 + d^6 + e^6)^{14} + 20(1 + a^3 + b^3 + c^3)^{30}(1 + d^3 + e^3)^{10} - (1 + a^2 + b^2 + c^2)^{30}(1 + d^2 + e^2)^{15} - 24(1 + a^{10} + b^{10} + c^{10})^6(1 + d^{10} + e^{10})^3 - 20(1 + a^6 + b^6 + c^6)^{10}(1 + d^6 + e^6)^5 - 15(1 + a + b + c)^4(1 + a^2 + b^2 + c^2)^{28}(1 + d + e)^{28} \right\} \quad (11)$$

The above multinomial generating functions can be expanded and simplified for each IR of the $I_h$ group, and these functions offer a greater flexibility for the edge colorings compared to the ordinary Pólya’s expansion, because the GF has terms partitioned into coloring hex–hex edges and pent–hex edges differently. Thus, the GFs obtained above have the ability to enumerate edge colorings with different specifications:

(a) if all hex–hex edges of $C_{60}$ are colored with one color (white) and pent–hex edges are colored with multiple colors;
(b) if all hex–hex edges are colored with multiple colors—say green, red, blue, burgundy, and cyan (see Figure 1b)—and all hex–pent edges are colored with white; this case also corresponds to coloring the 30 edges of the icosahedron as shown in Figure 1a, b.
(c) if both pent–hex and hex–hex edges are colored with multiple colors from a single set $R$ of colors without making any distinction between the two sets of edges.
(d) if all hex–hex edges are colored with colors from a single set, while all pent–hex edges are colored with colors chosen from a different set of colors, thus making a distinction between the pent–hex and hex–hex edges.

For each of the cases listed above, one can obtain a set of GFs for each IR of the $I_h$ group, thus yielding a powerful set of enumerative combinatorial generating functions. Hence, we consider each such case.

For case (a), the edge colorings of $C_{60}$ are obtained by setting $d$ and $e$ to 0 in Equation (10), which yields Equation (12):

$$GF_{G}^{A_g} = \frac{1}{120} \left\{ 1 + a + b + c \right\}^{60}(1 + a^2 + b^2 + c^2)^{30} + 24(1 + a^5 + b^5 + c^5)^{12}(1 + a^3 + b^3 + c^3)^{20} + (1 + a^2 + b^2 + c^2)^{30} + 15(1 + a + b + c)^4(1 + a^2 + b^2 + c^2)^{28} \right\} \quad (12)$$

when the above expressions are expanded and simplified for each IR, the results can be expressed in terms of partitions of integer 60 in 4 parts. For example, the partition $[15 15 15 15]$ of 60 corresponds to the term $1^{15}a^{15}b^{15}c^{15} = a^{15}b^{15}c^{15}$ and hence, the coefficient $a^{15}b^{15}c^{15}$ in the multinomial expansion gives the number of ways 60 pent–hex edges of $C_{60}$ can be colored with 15 white, 15 green, 15 blue, and 15 red colors keeping all hex–hex edges white. The $GF$ for the $A_u$ IR generates the number of chiral pairs for the corresponding chiral partition $[15 15 15 15]$ for the pent–hex colorings. Thus, the sum of the numbers for $A_g$ and $A_u$ enumerated for each color partition yields the total number of all possible pent–hex colorings that encompass both achiral colorings and chiral pairs. In general, for each IR of the $I_h$ group the results enumerated represent the number of colorings that transform according to the IR for the given color partition. These general results for any IR find applications to nuclear spin statistics beyond electrons, for example, spin 3/2 fermions and quarks [57–59].
The GF is obtained for the case \((b)\) by setting the edge weights \(a, b,\) and \(c\) to 0 in Equation (10), which results in Equation (13):

\[
GF^A_G = \frac{1}{120} \left\{ (1 + d + e)^{30} + 15 (1 + d + e)^{2} (1 + d^2 + e^2)^{14} + 24 (1 + d^3 + e^3)^{6} + 20 (1 + d^3 + e^3)^{10} + (1 + d^2 + e^2)^{15} + 24 (1 + d^{10} + e^{10})^{3} + 20 (1 + d^6 + e^6)^{5} + 15 (1 + d + e)^{4} (1 + d^2 + e^2)^{13} \right\}. \tag{13}
\]

Expression (13) thus obtained for the \(A_g\) IR yields the number of equivalence classes of coloring hex–hex edges with 3 different colors while keeping all the pent–hex colors white. The above expression when generalized to include two more colors with weights \(f\) and \(g\)—that is, every term \((1 + d^k + e^k)\) in Equation (13) is replaced with \((1 + d^k + e^k + f + g)\)—we obtain the results shown in Table 2 for the colorings of the edges of the icosahedron (Figure 1a,b) with up to 5 different colors under the action of the \(I_h\) group. Only some of the color partitions are shown in Table 2, as the number of terms in the multinomial GF for coloring 30 edges with 5 different colors is \(\binom{30+5-1}{30} = \binom{34}{30} = 46,376\).

If all edges are colored with colors chosen from a single set of colors (case \((c)\)), we obtain the GF for each IR by setting \(u_{ij} = u_i\) for all \(i\), which leads to the following GF for \(A_g\) for the case with 4 colorings of 120 edges of \(C_{80}\) (\(I_h\)) without differentiating the pent–hex and hex–hex edges. Thus, the expression obtained is shown in Equation (14):

\[
GF^A_G = \frac{1}{120} \left\{ (1 + a + b + c)^{120} + 15 (1 + a^2 + b^2 + c^2)^{60} + 24 (1 + a^3 + b^3 + c^3)^{12} + 20 (1 + a^3 + b^3 + c^3)^{10} + (1 + a^2 + b^2 + c^2)^{60} + 24 (1 + a^{10} + b^{10} + c^{10})^{12} + 20 (1 + a^6 + b^6 + c^6)^{20} + 15 (1 + a + b + c)^{8} (1 + a^2 + b^2 + c^2)^{56} \right\}. \tag{14}
\]

The above expression (14) can be expanded; except for the first few terms, there is a combinatorial explosion. Note that the number of terms in the above expansion or the number of ordered partitions of 120 into 4 parts is \(\binom{120 + 4 - 1}{120} = \binom{123}{120} = 302,621\). Consequently, an exhaustive construction of the combinatorial tables for all such edge colorings will take too much space, although they have all been generated by our computer code. Hence, we restrict to some of the binomial coloring terms shown in Tables 3–5.

However, for any given color partition, the respective coefficient can be directly obtained from the GFs without having to expand the entire multinomial function for the edge colorings. Thus, we have constructed the tables of our enumerations for the \(A_g\) and \(A_u\) IRs of the \(I_h\) group for \(C_{80}, C_{180},\) and \(C_{240}\) only for binomial colorings. Owing to a large number of edges for the giant fullerenes, both the number of terms in the expansion and the coefficients of various terms result in a combinatorial explosion for the edge colorings. Tables 3–5 show our computed results for the three giant fullerenes for only binomial colorings (black and white colors) for coloring the edges for the \(A_g\) and \(A_u\) IRs. Although as shown above, we can obtain a more itemized distribution for the edge coloring such as pent–hex, hex1–hex1, hex1–hex2 … colorings depending on the number of equivalence classes of hex–hex edges, for the sake of simplicity, we show in Tables 3–5 only the overall binomial color distributions without making further distinction into the types of edges. Table 3 shows the binomial colorings of 120 edges of \(C_{80}\), where in Table 3, \(k\) represents the number of black colors while 120–\(k\) is the number of white colors. We have shown the results for both \(A_u\) and \(A_g\) IRs where the numbers for \(A_u\) yield the numbers of chiral pairs for each edge-coloring partition shown in Table 3. As can be seen from Table 3, one needs a minimum of two black colors with the remaining 118 edges colored in white in order to produce a chiral edge coloring. As the number of black colors increases, the number of chiral colorings also increases,
reaching a maximum of 805,124,240,336,360,915,214,413,591,091,584 chiral pairs. The corresponding number for \( A_g \) is 805,124,240,336,361,157,749,996,742,071,252, suggesting that as we approach the peak of the binomial distribution, almost every edge coloring is chiral.

Table 2. Edge colorings of an icosahedron with up to 5 different colors (green, blue, red, cyan, burgundy)—one such coloring is in Figure 1.

| Color Partition | \( A_g \) | \( A_u \) |
|-----------------|---------|---------|
| 30 0 0 0 0      | 1       | 0       |
| 29 1 0 0 0      | 1       | 0       |
| 28 2 0 0 0      | 8       | 3       |
| 27 3 0 0 0      | 46      | 32      |
| 26 4 0 0 0      | 262     | 221     |
| 25 5 0 0 0      | 1257    | 1166    |
| 24 6 0 0 0      | 5113    | 4912    |
| 23 7 0 0 0      | 17,238  | 16,674  |
| 22 8 0 0 0      | 49,270  | 48,620  |
| 21 9 0 0 0      | 119,997 | 118,996 |
| 20 10 0 0       | 251,512 | 249,995 |
| 19 11 0 0       | 456,729 | 454,727 |
| 18 12 0 0       | 722,760 | 720,125 |
| 17 13 0 0       | 1,000,251 | 997,248 |
| 16 14 0 0       | 1,214,376 | 1,210,944 |
| 15 15 0 0       | 1,295,266 | 1,291,834 |
| 28 1 1 0 0      | 9       | 6       |
| 27 2 1 0 0      | 113     | 97      |
| 26 3 1 0 0      | 937     | 897     |
| 25 4 1 0 0      | 6019    | 5902    |
| 24 5 1 0 0      | 29,835  | 29,598  |
| 23 6 1 0 0      | 119,106 | 118,396 |
| 22 7 1 0 0      | 390,754 | 389,818 |
| 21 8 1 0 0      | 1,074,073 | 1,072,500 |
| 20 9 1 0 0      | 3,205,217 | 3,202,786 |
| 19 10 1 0       | 5,009,719 | 5,006,287 |
| 18 11 1 0       | 8,652,131 | 8,647,535 |
| 17 12 1 0       | 12,977,523 | 12,971,946 |
| 16 13 1 0       | 16,969,947 | 16,963,512 |
| 15 14 1 0       | 19,393,980 | 19,387,116 |
| 26 2 2 0 0      | 1438    | 1355    |
| 25 3 2 0 0      | 12,025  | 11,817  |
| 24 4 2 0 0      | 74,698  | 74,087  |
| 23 5 2 0 0      | 357,162 | 355,914 |
| 22 6 2 0 0      | 1,367,704 | 1,365,026 |
| 21 7 2 0 0      | 4,295,434 | 4,290,858 |
| 20 8 2 0 0      | 11,272,690 | 11,264,825 |
| 19 9 2 0 0      | 25,043,735 | 25,034,295 |
| 18 10 2 0       | 47,583,250 | 47,566,805 |
| 17 11 2 0       | 77,838,703 | 77,838,111 |
| 16 12 2 0       | 110,297,148 | 110,271,837 |
| 15 13 2 0       | 135,747,564 | 135,720,108 |
| 14 14 2 0       | 145,443,696 | 145,414,524 |
| 26 1 1 1 1      | 5484    | 5478    |
| 22 2 2 2        | 122,923,788 | 122,907,252 |
| 18 3 3 3        | 266,399,185,320 | 266,399,082,360 |
| 14 4 4 4        | 76,423,285,357,360 | 76,423,248,055,440 |
| 10 5 5 5        | 2,937,587,511,706,920 | 2,937,587,492,247,480 |
| 6 6 6 6        | 11,423,951,476,076,040 | 11,423,951,339,159,200 |

\(^a\) Numbers under \( A_u \) are chiral pairs of colorings and the sum of \( A_g \) and \( A_u \) is the total number of colorings and \( A_g - A_u \) is the number of achiral colorings for each color partition listed in the first column for coloring 30 edges of an icosahedron.
### Table 3. Edge colorings of C₈₀.

| n   | $A_n$ | $A_{n-1}$ |
|-----|-------|-----------|
| 11  | 1     | 0         |
| 120 | 1     | 0         |
| 119 | 2     | 0         |
| 118 | 78    | 56        |
| 117 | 3     | 2248      |
| 114 | 5     | 1586216   |
| 116 | 4     | 30434316  |
| 113 | 7     | 375768364 |
| 112 | 8     | 395608280 |
| 111 | 9     | 395608280 |
| 110 | 10    | 395608280 |
| 109 | 11    | 2673344820 |
| 108 | 12    | 2673344820 |
| 107 | 13    | 2673344820 |
| 106 | 14    | 2673344820 |
| 105 | 15    | 2673344820 |
| 104 | 16    | 2673344820 |
| 103 | 17    | 2673344820 |
| 102 | 18    | 2673344820 |
| 101 | 19    | 2673344820 |
| 100 | 20    | 2673344820 |
| 99  | 21    | 2673344820 |
| 98  | 22    | 2673344820 |
| 97  | 23    | 2673344820 |
| 96  | 24    | 2673344820 |
| 95  | 25    | 2673344820 |
| 94  | 26    | 2673344820 |
| 93  | 27    | 2673344820 |
| 92  | 28    | 2673344820 |
| 91  | 29    | 2673344820 |
| 90  | 30    | 2673344820 |
| 89  | 31    | 2673344820 |
| 88  | 32    | 2673344820 |
| 87  | 33    | 2673344820 |
| 86  | 34    | 2673344820 |
| 85  | 35    | 2673344820 |
| 84  | 36    | 2673344820 |
| 83  | 37    | 2673344820 |
| 82  | 38    | 2673344820 |
| 81  | 39    | 2673344820 |
| 80  | 40    | 2673344820 |
| 79  | 41    | 2673344820 |
| 78  | 42    | 2673344820 |
| 77  | 43    | 2673344820 |
| 76  | 44    | 2673344820 |
| 75  | 45    | 2673344820 |
| 74  | 46    | 2673344820 |
| 73  | 47    | 2673344820 |
| 72  | 48    | 2673344820 |
| 71  | 49    | 2673344820 |
| 70  | 50    | 2673344820 |
| 69  | 51    | 2673344820 |
| 68  | 52    | 2673344820 |
| 67  | 53    | 2673344820 |
| 66  | 54    | 2673344820 |
| 65  | 55    | 2673344820 |
| 64  | 56    | 2673344820 |
| 63  | 57    | 2673344820 |
| 62  | 58    | 2673344820 |
| 61  | 59    | 2673344820 |
| 60  | 60    | 2673344820 |
Table 4. Edge colorings of C\textsubscript{180}.

| k   | A\textsubscript{g} | A\textsubscript{u} |
|-----|----------------|------------------|
| 1   | 1              | 0                |
| 2   | 4              | 1                |
| 3   | 273            | 2686             |
| 4   | 1807,917       | 1803,450         |
| 5   | 96,020,482     | 95,988,421       |
| 6   | 4,240,271,777  | 4,240,025,284    |
| 7   | 159,913,119,758| 159,913,591,426 |
| 8   | 5,257,122,017,406| 5,257,112,226,096|
| 9   | 153,040,502,697,936| 153,040,448,776,803|
| 10  | 3,994,356,526,363,671| 3,994,356,224,947,792|
| 11  | 9,441,205,997,837,388| 9,441,205,809,631,423|
| 12  | 2,037,726,939,692,823,911| 2,037,726,932,160,044,746|
| 13  | 40,441,042,272,456,578| 40,441,042,232,722,631,201|
| 14  | 742,381,989,976,934,452| 742,381,989,780,438,407,424|
| 15  | 12,669,985,999,837,504,108| 12,669,985,968,812,183,074|
| 16  | 201,927,901,238,727,703,128| 201,927,901,235,891,706,182,400|
| 17  | 3,017,040,406,724,166,624,490,064| 3,017,040,406,712,915,379,645,072|
| 18  | 42,406,179,050,007,222,298,565,790| 42,406,179,049,962,711,561,688,894|
| 19  | 562,439,848,452,470,797,536,343,504| 562,439,848,452,304,546,365,120,176|
| 20  | 7,058,620,098,077,732,214,695,554,434| 7,058,620,098,077,113,629,220,104,300|
| 21  | 84,031,191,643,777,442,794,988,130,060| 84,031,191,643,777,256,084,971,286,460|
| 22  | 1,038,536,688,151,325,854,648,652,352,800| 1,038,536,688,151,324,923,452,261,432,364,005|
| 23  | 12,669,985,999,837,504,108,000| 12,669,985,968,812,183,074,000|
| 24  | 201,927,901,238,727,703,128,000| 201,927,901,235,891,706,182,400,000|
| 25  | 3,017,040,406,724,166,624,490,064,000| 3,017,040,406,712,915,379,645,072,000|
| 26  | 42,406,179,050,007,222,298,565,790,000| 42,406,179,049,962,711,561,688,894,000|
| 27  | 562,439,848,452,470,797,536,343,504,000| 562,439,848,452,304,546,365,120,176,000|
| 28  | 7,058,620,098,077,732,214,695,554,434,000| 7,058,620,098,077,113,629,220,104,300,000|
| 29  | 84,031,191,643,777,442,794,988,130,060,000| 84,031,191,643,777,256,084,971,286,460,000|
| 30  | 1,038,536,688,151,325,854,648,652,352,800,000| 1,038,536,688,151,324,923,452,261,432,364,005,000|

Table 4 shows our computed results as for the binomial edge colorings of C\textsubscript{180}. A striking contrast between C\textsubscript{180} and C\textsubscript{80} as well as C\textsubscript{60} is that the number of A\textsubscript{g} colorings for k = 1 are 4 for C\textsubscript{180}, which means there are 4 kinds of edges for C\textsubscript{180}, unlike the smaller icosahedral fullerenes. Moreover, there is exactly one chiral pair of edge coloring for the case k = 1 as inferred from Table 4 for the A\textsubscript{u} IR. There 5 edge colorings for C\textsubscript{180} with 269 white colors and 1 black color, among which there exists a chiral pair: a result that was not known until now. For the case of k = 2 or two black colors, it is seen that among 639 edge colorings for C\textsubscript{180}, there are 294 chiral pairs, and the remaining 51 colorings are totally achiral. Analogous to C\textsubscript{80}, the number of edge colorings grows combinatorially, resulting in an explosion already for k = 30, and hence, numerical results are not shown beyond k = 30. We see from Table 4 that almost all edge colorings of C\textsubscript{180} are chiral. Although we do not consider the case of C\textsubscript{140} icosahedral fullerene, the parent cage itself is chiral, as it belongs to the I group. Consequently, every edge coloring of C\textsubscript{140} is chiral, which is a direct consequence of the chirality of the parent cage. The results in Table 5 for the edge colorings of C\textsubscript{240}, the largest of the fullerenes considered here, reveal that there are 6 edge colorings for the one black color and the remaining white colors, among which there is exactly a chiral pair, and the remaining 4 colorings are achiral. The numbers for k = 2 (two black colors) suggest that there are 1122 edge colorings with 523 chiral pairs and 76 achiral colorings. As seen from Table 5, for k = 26 (26 black colors), there are 23,844,803,619,432,111,008,435,439,294,860,623,872 chiral edge colorings of edges and 74,228,417,154,323,763,200 achiral colorings for C\textsubscript{240}. 
The total numbers of all edge colorings, the number of chiral pairs, and the number of achiral colorings can be readily derived from the numbers enumerated in the tables for C_{80} through C_{240}. Let $N_k(A_g)$ and $N_k(A_u)$ be the numbers enumerated in the tables for the various values of $k$. Thus, we obtain:

\[ N_k \text{(Total; Binomial Edge colors)} = N_k(A_g) + N_k(A_u) \] (15)

\[ C_k \text{(Chiral; Binomial Edge colors)} = 2N_k(A_u) \] (16)

\[ AC_k \text{(Achiral; Binomial Edge colors)} = N_k(A_g) - N_k(A_u) \] (17)

We also note that as $k$ approaches $3n/4$, where $n$ is the number of vertices of the fullerene, we obtain Equation (18):

\[ \lim_{k \to 3n/4} \frac{2C_k}{2N_k(A_g) + N_k(A_u)} = 1. \] (18)

As can be seen from Tables 2–5, the numbers of edge colorings rise astronomically even for 2 colorings, and hence, we obtain analytical expressions for any $k$. Such expressions can be derived from the original GCCI for the $A_g$ and $A_u$ IRs for the edge colorings of the giant $C_{240}$ fullerene cage as shown below:

\[ N_k(A_g; C_{240}, k \text{ black edge colors}) = \frac{1}{120} \left( \binom{360}{k} + 15\eta \left( \binom{k}{2} \right) \left( \binom{180}{k/2} \right) + 24\eta \left( \binom{k}{5} \right) \left( \binom{72}{k/5} \right) + 
\right.

\[ + 20\eta \left( \binom{k}{3} \right) \left( \binom{120}{k/3} \right) + \eta \left( \binom{k}{10} \right) \left( \binom{180}{k/10} \right) + 24\eta \left( \binom{36}{k/10} \right) + 20\eta \left( \binom{k}{6} \right) \left( \binom{60}{k/6} \right) + 
\]

\[ 15\Sigma_{\lambda\lambda} \left( \binom{16}{\lambda} \right) \left( \binom{172}{k - \lambda} \right). \] (19)
where \( \eta\left(\frac{k}{3}\right) = 1 \) if \( d \) divides \( k \) and 0 otherwise, where \([\ ]\) is an ordered partition of \( k \) into 2 parts such that both parts must be even.

\[
N_k(A_g;C_{240}; k \text{ black edge colors}) = \frac{1}{120}\left(\frac{360}{k}\right) + 15\eta\left(\frac{k}{2}\right)\left[\binom{180}{k}\right] + 24\eta\left(\frac{k}{6}\right)\left[\binom{72}{k}\right] + 20\eta\left(\frac{k}{3}\right)\left[\binom{120}{k}\right] - \eta\left(\frac{k}{2}\right)\left[\binom{180}{k}\right] - 24\eta\left(\frac{k}{10}\right)\left[\binom{36}{k}\right] - 20\eta\left(\frac{k}{6}\right)\left[\binom{60}{k}\right] - 15\sum_{[\lambda]}
[
\left[\frac{16}{[\lambda]}\right]\left[\frac{172}{k - [\lambda]}\right]
]\right),
\]

where \( \eta\left(\frac{k}{2}\right) = 1 \) if \( d \) divides \( k \) and 0 otherwise.

We can also obtain the number of inequivalent ways of labeling all the edges of an icosahedral fullerene with \( n \) vertices as follows:

\[
N_{Lab}(I_h; C_n; \text{ fullerene with } n \text{ vertices}) = \frac{(3n/2)!}{120}. \tag{21}
\]

We can divide the above expression further into pent–hex and hex–hex edges with expressions (22) and (23), respectively:

\[
N_{Lab}(C_n I_h; \text{ pent – hex}) = \frac{60!}{120} \tag{22}
\]

\[
N_{Lab}(C_n I_h; \text{ hex – hex}) = \frac{(3n-120)!}{120}. \tag{23}
\]

4. Conclusions

In the present study, we have outlined powerful combinatorial techniques that synthesized Möbius inversion with the generalized version of Sheehan’s modification of Pólya’s theorem to all characters of the irreducible representations of the icosahedral group and applied the techniques to enumerate the edge colorings of giant fullerene cages. We developed a computer code that was used to construct the results specifically for the binomial chiral and achiral edge colorings of giant fullerenes. We also showed how the techniques can be divided into combinatorial parts with varied color partitions for the pent–hex and hex–hex edges contained in these giant fullerenes. The existence of chiral colorings even for one black edge coloring was demonstrated for the \( C_{180} \) and \( C_{240} \) fullerenes. It was shown that the ratio of chiral colorings and total number of edge colorings asymptotically reach unity as the number of black colors becomes almost equal to the number of white colors. The present enumeration of edge colorings, especially with restrictions to nearest neighbor exclusion, can be of immense use in the combinatorics of equivalence classes of Kekule structures and thus in the computations of stabilities of giant fullerenes using the conjugated circuit method or the topological resonance theory method. The present techniques considered in this study could have applications to NMR, multiple quantum NMR, and ESR where one seeks the number of different types of dipolar couplings or edge colorings. The present enumeration techniques can also be extended in the future to nanomaterials such as carbon nanotubes, carbon nanocones, nanotori, and other nanomaterials of ongoing experimental interest.

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