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The Total π-Electron Energy Saga

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Abstract: The total π-electron energy, as calculated within the Hückel tight-binding molecular orbital approximation, is a quantum-theoretical characteristic of conjugated molecules that has been conceived as early as in the 1930s. In 1978, a minor modification of the definition of total π-electron energy was put forward, that made this quantity interesting and attractive to mathematical investigations. The concept of *graph energy,* introduced in 1978, became an extensively studied graph-theoretical topic, with many hundreds of published papers. A great variety of graph energies is being considered in the current mathematical-chemistry and mathematical literature. Recently, some unexpected applications of these graph energies were discovered, in biology, medicine, and image processing.

We provide historic, bibliographic, and statistical data on the research on total π-electron energy and graph energies, and outline its present state of art. The goal of this survey is to provide, for the first time, an as-complete-as-possible list of various existing variants of graph energy, and thus help the readers to avoid getting lost in the jungle of references on this topic.

Keywords: total π-electron energy, graph energy, molecular graph, chemical graph theory, spectral graph theory.

INTRODUCTION

THE total π-electron energy (E_π), as calculated by the simplest tight-binding approximation of the molecular orbital theory, appeared for the first time in the early 1930s, in Erich Hückel's seminal paper[1] "*Quantentheoretische Beiträge zum Benzolproblem I. Die Elektronenkonfiguration des Benzols und verwandter Verbindungen*" in which it was shown that in the case of benzene

$$E_\pi = 6\alpha + 8\beta$$

with α and β representing, respectively, the Coulomb integral pertaining to a carbon atom and the carbon-carbon resonance integral. The method that eventually became known as the Hückel molecular orbital (HMO) theory enabled a reasonably accurate quantitative prediction of thermodynamic properties, resonance stabilization, and aromaticity of conjugated polycyclic π-electron systems and gained much popularity in the next 50 years.[2–4] A bibliography of the early researches of total π-electron energy[5] and details of its chemical applications can be found in the surveys[6,7] and books.[8,9]

The connection between HMO theory and spectral graph theory is nowadays a well established and fully elaborated part of chemical graph theory.[8–11] If the eigenvalues of a molecular graph (of a conjugated π-electron system) are labeled as $\lambda_1 \geq \lambda_2 \geq \cdots \geq \lambda_n$, then the energy of the *i*-th molecular orbital is

$$E_i = \alpha + \lambda_i \beta .$$

Within the HMO approximation, the total π-electron energy is then equal to

$$E_\pi = \sum_{i=1}^{n} g_i E_i$$

where g_i is the number of π-electrons in the *i*-th molecular orbital. For uncharged conjugated hydrocarbons,

$$\sum_{i=1}^{n} g_i = n$$

where *n* is the number of vertices of the underlying molecular graph. For such molecules in their ground electronic state, if $n = 2k$, then

$$g_1 = g_2 = \cdots = g_k = 2 \ , \ g_{k+1} = g_{k+2} = \cdots = g_n = 0$$

whereas if $n = 2k + 1$, then

$$g_1 = g_2 = \cdots = g_k = 2,\ g_{k+1} = 1,\ g_{k+2} = g_{k+3} = \cdots = g_n = 0.$$

Consequently,

$$E_\pi = \begin{cases} n\alpha + \beta\left[2\sum_{i=1}^{n/2} \lambda_i \right] & \text{if } n \text{ is even} \\ n\alpha + \beta\left[\lambda_{(n+1)/2} + 2\sum_{i=1}^{(n-1)/2} \lambda_i \right] & \text{if } n \text{ is odd} \end{cases}$$

The only non-trivial part of this expression is what formally is obtained by setting $\alpha = 0$ and $\beta = 1$, usually referred to as the total π-electron energy in β-units:

$$E_\pi = \begin{cases} 2\sum_{i=1}^{n/2} \lambda_i & \text{if } n \text{ is even} \\ \lambda_{(n+1)/2} + 2\sum_{i=1}^{(n-1)/2} \lambda_i & \text{if } n \text{ is odd} \end{cases} \quad (1)$$

The opinion of one of the present authors (who in 1978 was somewhat younger than nowadays) was that mathematicians will hardly ever be interested to study a graph-spectrum-based quantity equal to the right-hand side of Equation (1). Therefore, in order to make total π-electron energy more attractive to the mathematical community, he made a seemingly absurd and scientifically risky step, and proposed that instead of E_π, Equation (1), attention be paid to the quantity defined as

$$E(G) = \sum_{i=1}^{n} |\lambda_i| \quad (2)$$

which he named *graph energy*.

The first paper in which graph energy was defined as the sum of absolute values of the eigenvalues of the (0,1)-adjacency matrix of a graph G, namely as Equation (2), appeared in 1978.[12] It is published (in English language) in a difficult-to-find journal whose full title is *Berichte der Mathematisch-Statistischen Sektion im Forschungszentrum Graz*.

The idea to define $E(G)$ as in Equation (3) and to name it "*energy*" has the following chemical justification.

The eigenvalues of many molecular graphs satisfy the condition

$$E_\pi = \begin{cases} \lambda_{n/2} \geq 0 \geq \lambda_{n/2+1} & \text{if } n \text{ is even} \\ \lambda_{(n+1)/2} = 0 & \text{if } n \text{ is odd} \end{cases} \quad (3)$$

In the 1970s this was a well known fact. For such graphs, recalling that the sum of all eigenvalues is equal to zero, it is elementary to verify the following:

***Proposition* 1.** If conditions (3) are obeyed, then the total π-electron energy, Equation (1), satisfies the relation

$$E_\pi = \sum_{i=1}^{n} |\lambda_i| .$$

The (implicit) awareness of Proposition 1 can be recognized in the works of Charles Coulson,[13] George Hall,[14] Klaus Ruedenberg,[15] Bernard McClelland,[16] and probably other pioneers of HMO theory.

While studying HMO theory, one of the present authors noticed that two important earlier discovered results, namely Coulson's integral formula[13]

$$E_\pi = \frac{1}{\pi}\int_{-\infty}^{+\infty}\left[n - \frac{ix \succ '(G,ix)}{\succ (G,ix)} \right] dx$$

and McClelland's inequality[16] $E_\pi \leq \sqrt{2mn}$ hold if and only if the conditions (3) are satisfied. In other words, the actual results of Coulson and McClelland were the following two results described below in Propositions 2 and 3.

***Proposition* 2.**[13] Let G be an arbitrary graph of order n, let $\succ (G,\lambda)$ be its characteristic polynomial, and $i = \sqrt{-1}$. Then

$$\sum_{i=1}^{n} |\lambda_i| = \frac{1}{\pi}\int_{-\infty}^{+\infty}\left[n - \frac{ix \succ '(G,ix)}{\succ (G,ix)} \right] dx.$$

***Proposition* 3.**[16] Let G be an arbitrary graph with n vertices and m edges. Then,

$$\sum_{i=1}^{n} |\lambda_i| \leq \sqrt{2mn}.$$

The observations stated above as Propositions 2 and 3 were the prime motivation to move from the mathematically repelling expressions (1) for HMO total π-electron energy to the much simpler expression (2). By means of this change, the previous HMO results (*i.e.*, Coulson's formula and McClelland's inequality for E_π) would anyway remain valid for the vast majority (but not all!) chemically interesting cases. It is worth noting that $E(G)$ coincides with E_π for all alternant systems (including acyclic and benzenoid), but differs from E_π in the case of fullerenes and nanotubes.

Other mathematical reasons why Equation (2) should be preferred over Equation (1) are explained in the book.[17]

Formula (2) is well defined for all graphs, and thus can be applied to all graphs, without the (mathematically frustrating) restriction to molecular graphs. This gives full freedom to researchers, and enables them to arrive at results unimaginable to those thinking in terms of "molecular graphs".

By introducing the concept of graph energy,[12] its author hoped that it will attract the attention of mathematicians, and that the Coulson and McClleland formulas will be just the first in a long series of exact and non-trivial mathematical results for $E(G)$ to be discovered.

This indeed happened, but more than a quarter-of-century later.

Table 1. Number of papers on graph energies published in the last twenty years, a total of over 760 papers. In the last few years, such papers were produced faster than one per week (o.p.w.). In the analogous table,[23] pertaining to the situation in March 2016, the total count of papers was around 630. Based on these data, an attenuation of graph-energy research is not to be expected in the foreseen future. The authors of this table are aware that there must be numerous additional papers, especially those published in India and China, that are not accounted for

year	papers	comment	year	papers	comment
1996	2		2007	34	
1997	0		2008	55	>o.p.w.
1998	2		2009	68	>o.p.w.
1999	6		2010	68	>o.p.w.
2000	4		2011	57	>o.p.w.
2001	12		2012	59	>o.p.w.
2002	3		2013	58	>o.p.w.
2003	5		2014	69	>o.p.w.
2004	9		2015	98	>o.p.w.
2005	15		2016	80	>o.p.w.
2006	11		2017	51	as in April

Already before the publication of the paper,[12] a few results that pertain to the energy of trees were obtained.[18] The paper[12] was followed by several attempts to popularize the graph-energy concept,[9,19–21] but the mathematical and mathematico—chemical community remained uninterested until the beginning of the 21-st century. This silence lasted some 25 years.

Then, however, a dramatic change happened, and almost suddenly, a large number of colleagues, from unrelated and geographically very distant places, started to study graph energy. The main goal of the present survey is to provide, for the first time, an as-complete-as-possible list of various existing variants of graph energy, and thus help the readers to avoid getting lost in the jungle of references on this topic. We hope that at least some of our chemist colleagues will find it interesting to see how the quantum-chemical concept of molecular-orbital total π-electron energy evolved into a variety of mathematically conceived "graph energies", and how these "graph energies" recently found surprising and unexpected applications. In the subsequent section, we begin our considerations by providing some statistical data on the current research of graph energy.[22,23]

STATISTICS

The data outlined in this section pertain to the state of research on graph energy as in April 2017 (as known to the authors). An analogous statistical analysis, corresponding to the situation in March 2016 can be found elsewhere.[23] As seen below, the progress achieved in just a single year is remarkable.

Table 2. Number of scholars from various countries who authored or coauthored at least one article on graph energy in the period 1996–2017 (as on April 2017). Their true count is somewhat greater because we did not distinguish between scholars with the same surname and different names beginning with the same letter. Thus, Xia Li, Xuechao Li, and Xueliang Li were counted as one. Note that in the analogous table,[23] pertaining to the situation in March 2016, Argentina, Israel, Kuwait, Malaysia, Malta, Poland, Taiwan, and Uruguay were not yet represented

country	no	country	no	country	no	country	no
Argentina	5	Germany	9	Malta	1	Slovenia	1
Australia	5	Greece	2	Mexico	2	South Africa	4
Austria	1	Hungary	2	Norway	1	South Korea	11
Belgium	2	India	140	Netherlands	4	Spain	2
Brazil	11	Indonesia	1	Pakistan	10	Sweden	1
Canada	9	Iran	51	Poland	1	Taiwan	1
Chile	11	Ireland	1	Portugal	2	Thailand	2
China	217	Israel	1	Romania	2	Turkey	11
Colombia	8	Italy	9	Russia	1	UK	8
Croatia	4	Japan	2	Saudi Arabia	3	Uruguay	2
France	3	Kuwait	3	Serbia	32	USA	47
Georgia	1	Malaysia	3	Slovakia	3	Venezuela	8

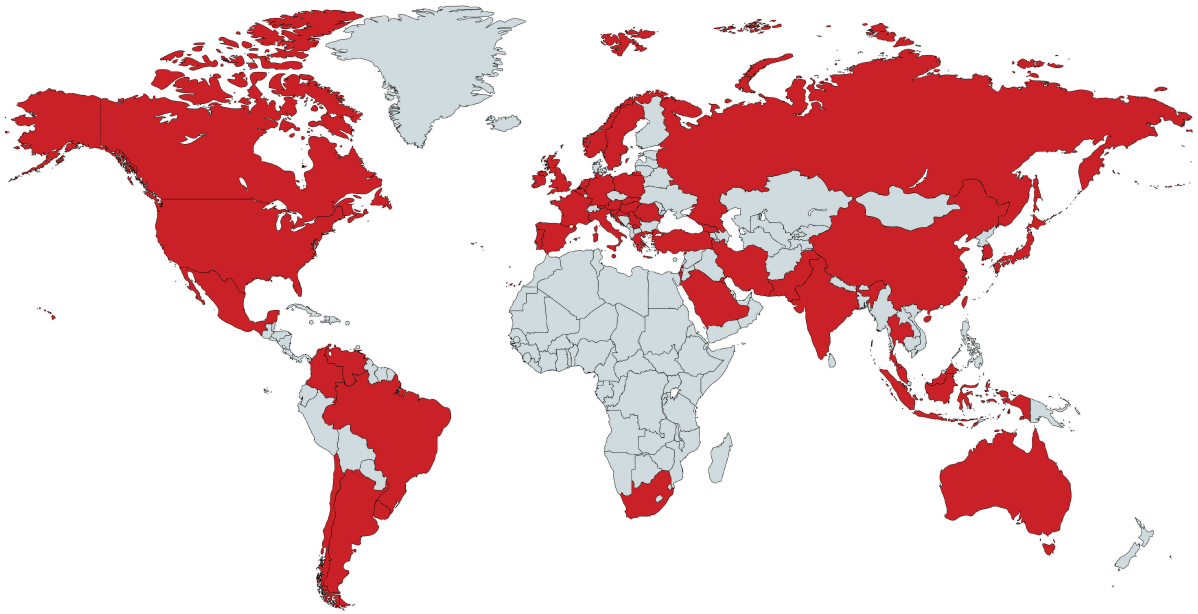

Figure 1. Countries where researches on graph energy were conducted (as by April 2017).

Somewhere around the year 2007, the number of publications on graph energy started to increase significantly. This trend is illustrated in Table 1.

Table 2, Figures 1 and 2 show the distribution of authors of graph-energy-papers by the affiliation countries. Figure 1 points out the countries in which these authors were employed, when creating their graph-energy articles.

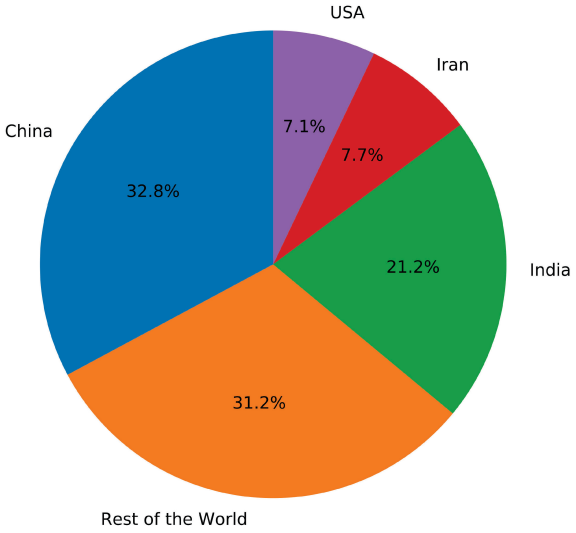

Figure 2. Relative number of authors by countries where they have been working in the time when their graph-energy articles were produced. This pie-chart indicates countries where the percentage of authors is greater than 5 %.

Figure 2 indicates the relative number of authors of graph-energy-papers by their affiliation countries.

The bibliography on which the data in Tables 1 and 2 are based, has been compiled by the authors of this survey. They made it as complete as they could, but it certainly is not 100 % complete. This implies that in reality, the count of articles and authors, stated in Tables 1 and 2, is even greater. It also may be that some more countries would need to be added to Table 2.

It should be noted that the mentioned bibliography does not include papers that are not directly related to graph energies. Thus, among others, not included into our statistics are the papers concerned with:

a) the countless approximate formulas for total π-electron energy (in terms of *n, m,* and other graph parameters), in particular such formulas for E_π of benzenoid hydrocarbons;[6,7]

b) empirical correlations between E_π and various structural parameters, in particular the Kekulè structure count;[24]

c) various resonance energies;[25,26]

d) studies of the energy effect of individual cycles in polycyclic conjugated molecules.[27,28]

THE GRAPH ENERGY DELUGE

Graph energy, Equation (2), is based on the eigenvalues of the ordinary (0,1)-adjacency matrix of the graph *G*. Its

mathematical examination resulted in a plethora of newly established properties and, consequently, in a plethora of published papers.[29] In view of this success, a natural idea was to look for some variant of graph energy that would also provide a basis for prolific mathematical researches.

The first such new graph energy was introduced as early as in 1994 by Yang, Xu and Hu.[30] They constructed the "extended adjacency matrix" whose (i,j)-element is equal to

$$\frac{1}{2}\left(\frac{d_i}{d_j}+\frac{d_j}{d_i}\right)$$

for adjacent vertices, and is zero otherwise; d_i stands for the degree of the i-th vertex. The respective "extended graph energy" did not attract any attention of other scholars and fall into oblivion. It was re-discovered only quite recently,[31–33] when a few of its properties were established.

The next step in this direction was to employ eigenvalues of another popular graph matrix. The most obvious candidate for this was the Laplacian matrix.[34,35] Let its eigenvalues be denoted by $\mu_1,\mu_2,\ldots,\mu_n$. Then, in analogy with Equation (2) one could conceive the Laplacian energy of the graph G as $LE(G)=\sum_{i=1}^{n}|\mu_i|$. However, because all Laplacian eigenvalues are non-negative, and because their sum is equal to $2m$, we would arrive at the trivial result $LE(G)=2m$. The way out of this difficulty was found by defining the Laplacian energy as[36]

$$LE(G)=\sum_{i=1}^{n}\left|\mu_i-\frac{2m}{n}\right| \qquad (4)$$

The Laplacian energy was the first in a long series of energies based on other graph matrices. It was followed by the distance energy (based on the eigenvalues of the distance matrix),[37] normalized Laplacian energy (based on the eigenvalues of the normalized Laplacian matrix[38] which independently was introduced under the name of "Randić energy"),[39] *etc.* Consonni and Todeschini[40] defined the energy of any real symmetric matrix with eigenvalues $x_1,x_2,\ldots,x_n$ as $\sum_{i=1}^{n}|x_i-S/n|$ where $S=x_1+x_2+\cdots+x_n$.

Nikiforov extended the energy-concept to any matrix.[41] If M is a $p\times q$ matrix (where p and q need not be equal), then the positive square roots of the eigenvalues of MM^t are the singular values of M. Here M^t denotes the transpose of the matrix M. According to Nikiforov, the energy of the matrix M is the sum of its singular values. In the case of real and symmetric (square) matrices, the new and the old energy-concepts coincide.

An "energy" can be associated also with polynomials.[42] Among such polynomial-based energies the "matching energy" plays an outstanding role.[43] In an implicit manner, this energy was conceived already in the 1970s,[44] and played role as a component of the so-called "topological resonance energy".[26,44–46]

At this point it is worth recalling that there are countless graph-based matrices.[47,48] Consequently, it is possible to imagine countless graph energies. In the last few years, this fact is misused by some scholars (most of which from India), who are introducing novel "graph energies" ad libitum, without any visible scientific justification. This phenomenon may be referred to as the "*graph energy deluge*", and should be viewed as a pathological feature of contemporary Mathematical Chemistry.

In our records, we have data on more than 90 different graph energies. In what follows, we give a list thereof, ordered according to the time of their first occurrence in the literature, with reference to the place where these have been considered for the first time. In reality, the number of existing graph energies may be still greater, and more such will for sure appear in the future.

1) (ordinary) graph energy[12]
2) extended adjacency energy[30]
3) Laplacian energy[36]
4) energy of matrix[41]
5) minimum robust domination energy[49]
6) energy of set of vertices[50]
7) distance energy[37]
8) Laplacian-energy-like invariant[51]
9) Consonni-Todeschini energies[40]
10) energy of (0,1)-matrix[52]
11) incidence energy[53]
12) maximum-degree energy[54]
13) skew Laplacian energy[55]
14) oriented incidence energy[56]
15) skew energy[57]
16) Randić energy[39]
17) normalized Laplacian energy[38]
18) energy of matroid[58]
19) energy of polynomial[42]
20) Harary energy[59]
21) sum-connectivity energy[60]
22) second-stage energy[61]
23) signless Laplacian energy[62]
24) *PI* energy[63]
25) Szeged energy[64]
26) He energy[65]
27) energy of orthogonal matrix[66]
28) common-neighborhood energy[67]
29) matching energy[43]
30) Seidel energy[68]
31) ultimate energy[69]
32) minimum-covering energy[70]
33) resistance-distance energy[71]
34) Kirchhoff energy[72]

35) color energy[73]
36) normalized incidence energy[74]
37) Laplacian distance energy[75]
38) Laplacian incidence energy[76]
39) Laplacian minimum dominating energy[77]
40) minimum-domination energy[78]
41) minimum-covering distance energy[79]
42) degree sum energy[80]
43) domination energy[81]
44) general Randić energy[82]
45) Randić incidence energy[83]
46) Laplacian minimum-covering energy[84]
47) minimum dominating distance energy[85]
48) *e*-energy[86]
49) *n*-energy[86]
50) double dominating energy[87]
51) Hermitian energy[88]
52) minimum hub distance energy[89]
53) minimum monopoly energy[90]
54) minimum monopoly distance energy[91]
55) complementary dominating energy[92]
56) upper dominating energy[93]
57) minimum-maximal-domination energy[94]
58) minimum-covering color energy[95]
59) alpha-distance energy[96]
60) alpha-incidence energy[96]
61) *so*-energy[97]
62) color Laplacian energy[98,99]
63) reciprocal complementary distance energy[100]
64) non-common neighborhood energy[101]
65) partition energy[102]
66) minimum equitable color dominating energy[103]
67) Nikiforov energy[104]
68) resolvent energy[105]
69) Laplacian resolvent energy[106]
70) signless Laplacian resolvent energy[106]
71) skew Randić energy[107]
72) geometric-arithmetic energy[108]
73) minimum hub energy[109]
74) (two) reduced color energies[110]
75) *o*-energy[111]
76) Co-*PI* energy[112]
77) Coxeter energy[113]
78) minimum dominating maximum degree energy[114]
79) minimum covering Seidel energy[115]
80) additive color Laplacian energy[116]
81) net-Laplacian energy[117]
82) Hermitian-Randić energy[118]
83) eccentric Laplacian energy[119]
84) *ABC* energy[120]
85) iota energy[121]
86) minimum boundary dominating energy[122]
87) Laplacian minimum boundary dominating energy[122]
88) minimum dominating
89) color signless Laplacian energy[124]
90) path energy[125]

By saying that the above list represents a "*pathological feature of mathematical chemistry*", the authors have expressed their personal critical opinion on this matter. On the other hand, the fact that the mentioned variants of graph energy do exist, cannot be and should not be ignored. A critical analysis of the advantages/disadvantages of these "graph energies" has not been done so far, and is not the purpose of the present survey. There is little doubt that in most cases, the authors proposing such graph invariants have not taken the trouble of not only motivating their new quantities but also of finding at least one application. Yet, some application of graph energies, other than in the HMO model, have been discovered, and are mentioned in a subsequent section.

BOOKS AND REVIEWS

At the present moment there are only two monographs entirely devoted to graph energies.[29,126] However, topics related to graph energy can be found in numerous other books and textbooks, sometimes in the form of entire chapters, sometimes mentioned only in a few lines.[8,9,17,127–145] There are also several reviews devoted to special topics of the theory of graph energy.[146–167]

APPLICATIONS OF GRAPH ENERGIES

In addition to its standard chemical application (based on its close relation with the HMO total π-electron energy),[2,3,6,8–10,25,28,168] the ordinary graph energy, Equation (2), and some other graph energies found unexpected applications in other areas of science.

Use of $E(G)$ in QSPR/QSAR studies was attempted on several occasions.[169,170] In particular, graph energies have been related to entropy.[171,172]

Properties of proteins (especially those of biological relevance) were modeled by means of graph-energy-like quantities in a number of recent papers.[173–177] There were attempts to use graph energy in the search for the genetic causes of Alzheimer disease[178] and for modeling of the spread of epidemics.[179] An unusual and somewhat outlandish biochemical use of graph energy is proposed in the paper,[180] whose title is "*Disruption of cell wall fatty acid biosynthesis in Mycobacterium tuberculosis using the concept of minimum robust domination energy of graph*".

The Laplacian energy, Equation (4), found a fully different and absolutely unforeseen area of application: in pattern recognition and image analysis,[181–186] which, in addition, is attempted to be used in medical investigations of brain activity.[187,188] The title of the paper[184] is characteristic: "*High-resolution satellite image classification and segmentation using Laplacian graph energy*".

These various biochemical, technical, and medicinal applications may serve as a justification of the abstract, purely academic, and seemingly arbitrary mathematical considerations by which the graph energies have been conceived. Such things often happen in science.

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