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The Work of Carlo Garosi: a Parallel History of the Discovery of Tritium

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Abstract

The advent of "New Physics" at the beginning of the XX century ignited a transformative era for Nuclear Physics, characterized by a dramatic growth of its knowledge and boundaries. The existence of isotopes, the discovery of neutrons and the advancement of experimental techniques all conjured groundbreaking discoveries, where experimental physicists were at the forefront of developments. The discovery of tritium, the hydrogen isotope of mass number 3, is among the most sought-after results in the 1930s, and we present evidence of how Carlo Garosi, an Italian pharmacist, made significant yet unacknowledged contributions in 1936. By recognizing the importance of the Oliphant, Harteck and Rutherford experiment of 1934, Garosi correctly identified the heavy atom discovered as an isotope of hydrogen.

This paper begins with the recapitulation of both the philosophical and scientific roots of his theory, showing how he critically engaged with older theories, such as Prout's 19th-century atomic theory, and integrated more recent works from international sources in original ways. Garosi proposed a unique interpretation of the Periodic Table, engaging in a critical review of the notion of how Helium is a result of the fusion of 4 Hydrogen atoms, and he explains the progression from one chemical element to another in terms of interactions among hydrogen, deuterium and tritium. By revisiting this seemingly peripheral event, we underscore its broader significance as a testament to the resilience and ingenuity of scientists journeying amid rising geopolitical tensions.

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Contents

1 Introduction

In 1934 at the Cavendish Laboratory in Cambridge, in series of experiments involving heavy water and *heavy hydrogen* Mark Oliphant, Paul Harteck and Ernest Rutherford observed an anomalous emission of protons, neutrons and other weak group of particles by bombarding deuterated ammonium chloride, ammonium sulphate and orthophosphoric acid with deuterons (or *diplons*, the chosen name at that time). As reported in their letter to the Editor of Nature [1]:

> While it is too early to draw definite conclusions, we are inclined to interpret the results in the following way [. . .] It seems more probable that the diplons unite to form a new helium nucleus of mass 4.0272 and 2 charges. This nucleus apparently finds it difficult to get rid of its large surplus energy above that of an ordinary He nucleus of mass 4.0022, but breaks up into two components. One possibility is that it breaks up according to the reaction $D_1^2 + D_1^2 \to H_1^3 + H_1^1$[1].

This is the first experimental evidence of the existence of tritium [2], the hydrogen isotope with a mass number of 3. Unfortunately, they were not able to isolate it, and as they stated [1]:

[1]We reported the original notation used by the authors for the nuclear charge number and the mass number.

> While the nuclei of H_1^3 and He_1^3 appear to be stable for the short time required for their detection, the question of their permanence requires further consideration.

A few months later, in a work published in the Proceedings of the Royal Society of London [3] Oliphant, Harteck and Rutherford reported a new series of experiments that provided further confirmation of the existence of an hydrogen isotope of mass 3. They proposed the following reaction:

$$_1D^2 +_1 D^2 \rightarrow_2 He^4 \rightarrow_1 H^1 +_1 H^3 \tag{1}$$

assuming that the first step in the reaction between two diplons is the formation of an excited helium nucleus. Then, on a very short timescale, the high-energy helium breaks up into a proton and a hydrogen isotope of mass 3. From energy and range calculations, paired with the analysis of the oscillograph deflections magnitude, the authors concluded that[3]:

> It seems clear that the production of this isotope of hydrogen of mass 3 in these reactions is established beyond doubt. The mass of the $_1H^3$ atom is consistent with its possessing a stability of the same order as $_1H^2$. The possible existence of this isotope has been discussed by several writers and although a careful search has been made no evidence of its presence has been found. It seems probable, however, that it could be formed by the process we have considered in sufficient quantity to be detected ultimately by spectroscopic or positive-ray methods.

The conclusive evidence of the existence of tritium (this name was originally proposed by Harold Urey, George Murphy and Ferdinand Brickwedde in 1933 [1]) was provided in 1939 by Luis Alvarez and Robert Cornog, who converted a cyclotron into a mass spectrometer dedicated to detect species with mass number 3 [4]. In a series of experiments conducted at the Radiation Laboratory in Berkeley they were able to demonstrate the stability of He^3 and the radioactivity of H^3. It is curious to note that in his initial investigations Rutherford made a serious mistake by considering tritium as the stable species and helium-3 as the radioactive one [5]. He firmly defended his idea and sought to convince other eminent scientists, like F.W. Aston, the inventor of the mass spectrometer, until the discovery of Alvarez

and Cornog compelled him to revise his stance.
This is the official historical reconstruction of tritium discovery. But there is another interesting story about its identification, that runs parallel to the experiments of Oliphant, Harteck and Rutherford.

2 An Independent Story

This alternative story begins in the small town of San Quirico d'Orcia, an ancient village in the countryside of Siena, en route on the Via Francigena, the old pilgrimage axis between Canterbury and Rome. The year is 1938, Italy is ruled by the Fascist dictatorship, and the ghosts of a new war in Europe are looming over the future. The regime decisions have long set aside the Italian scientific community from its counterparts in Europe and beyond, and lack of funds, small scientific institutions, and isolationism have thwarted the progress in the new branches of physics, especially nuclear physics. However, a genuine interest propels the debate among a community which is not restricted only to academia. This larger audience, albeit geographically far from the vanguard, avidly follows the latest developments.

Figure 1: *Photo of Carlo Garosi, courtesy of Venera Dibilio*

Carlo Garosi [San Quirico d'Orcia, 05/07/1883 - Florence, 22/12/1954], a pharmacist in San Quirico d'Orcia, is part of this community. Figure 1 is a photography of him, courtesy of his descendants. The connections between pharmacy and chemistry, both on historical and scientific grounds, are well known, and it is not surprising that a member of the order kept up with the news. The Authors had the opportunity to work with Garosi's personal archive. The archive has not been properly indexed yet, and a conservation effort is required to allow the scientific community to access it. Garosi subscribed to many publications, and he was also the author of a number of articles ([6] - [10]), on national magazines like "*Chimica. Rivista Mensile per la Diffusione della Cultura Chimica*"[2],

[2] "*Chemistry. Monthly Journal for the Promotion of Chemical Culture*", tr. by the Authors

published by the "*Istituto Italiano di Storia della Chimica*"[3] on the topic of fundamental components of matter. In July 1937 Garosi begins the publication of his magnum opus ([11] - [14]), where he recollects all the evidences - mainly theoretical - supporting the existence of an elementary constituent, tritium, two years before the experimental confirmation by Alvarez and Cornog [4]. Figure 2 shows the typewritten edition of his paper. Garosi proved to be a fine expert of chemistry, reviewing and building on a theory that dates back to 1815.

2.1 An Old Theory Resumed

William Prout (1785–1850), an English chemist, noting how element masses seemed to be multiples of the hydrogen weight, hypothesized in 1815 that all the nuclei were constituted of fundamental entities, called *protyles*, the hydrogen nucleus [15] [16]. His theory showed some serious flaws [17], because of "anomalies" such as *chlorine* of atomic weight 35.45, and it was definitely abandoned following the discovery of the neutron by Chadwick in 1932 ([18] - [20]). However, the concept of *isotope* allowed Garosi to update Prout's theory, and in 1937 he published an article *"Teoria sulla genesi degli elementi chimici"*[4] [11], in which he claimed the development of a comprehensive theory that linked all existent chemical element to a combination of H^1, H^2 or H^3.

2.2 Prout's Hypothesis

A quick review of the original theory is needed before we introduce Garosi's advancements. William Prout was an English physician at a time when the borders between chemistry and physiology were neither established nor understood. His major scientific achievement was the discovery of urea in urine [21], and its composition, alongside the discovery of hydrochloric acid in gastric juices of animals [22]. His scientific interests spanned from urinary and digestive pathologies to atomic theory. During his time the debate between vitalism and a materialistic and mechanistic view took a turn toward chemistry and a quantitative approach to physiology. Being interested into the chemical composition of organic fluids, he approached the

[3] "*Italian Institute for the History of Chemistry*", translation by the Authors

[4] *"Theory on the Origin of Chemical Elements"*, translation by the Authors

matter studying meticulously the weight of the compounds involved in his analysis. His hypothesis suggests that hydrogen might be the foundational "brick" of all elements, whose atomic mass would be an integer multiple of hydrogen's, that Prout names *protyle*, a Greek name from *prōt-*, proto, and *hylē*, substance. This idea contributed to the scientific discussions about the atomic theory, but failed to explain anomalies such of chlorine and other elements, and was later discarded.

3 Carlo Garosi's Original Work

3.1 Genesis of a Theory

The scientific environment at Garosi's time is very different from the one Prout experienced. Quantum Mechanics and General Relativity have made their appearance on the global stage, and they have quickly become mainstream, although not unchallenged [23]. Differences in the established atomic weight were now explained also through concepts such as energy binding, and the development of mass spectroscopy won the Nobel Prize for F. W. Aston in 1922, with the discovery of isotopes. An echo of Prout's hypothesis reverberates through the motivation of Aston's prize [24]:

> [...] for his discovery, by means of his mass spectrograph, of isotopes, in a large number of non-radioactive elements, and for his enunciation of the whole-number rule.

The discovery of neutron by Chadwick in 1932 [19] complicated things, although its status within the atomic nuclei was uncertain. In this early stage of nuclear physics, where the influence of experiments was quickly rewriting how physicists were supposed to develop hypothesis, Carlo Garosi completed his reorganization of the table of elements. His hypothesis is shaped by two main general principles, which are interesting to highlight the philosophical roots of his work:

Unitary principle: Garosi draws inspiration from the "alchemic tradition", particularly its philosophical assertion that all matter originates from a single, unified source and transforms into diverse elements through processes of change. This concept is encapsulated in the phrase "The whole is one", which he prominently cites at the beginning of his paper[5]. This principle serves as a foundation for his interpretation of the genesis and progression of chemical elements;

Evolutionary principle: through his work, Garosi applies a synthesis of evolution and Aristotelian principles, trying to frame evolution as depending on "intelligent design from within" and "environmental pressure from outside"[6], and applying this scheme to both biology and chemistry: chemical elements are both "prone" to react with different compounds based on their properties and also because of the environment they happen to exist in, due to time and other factors.

These two principles account for the idea that:

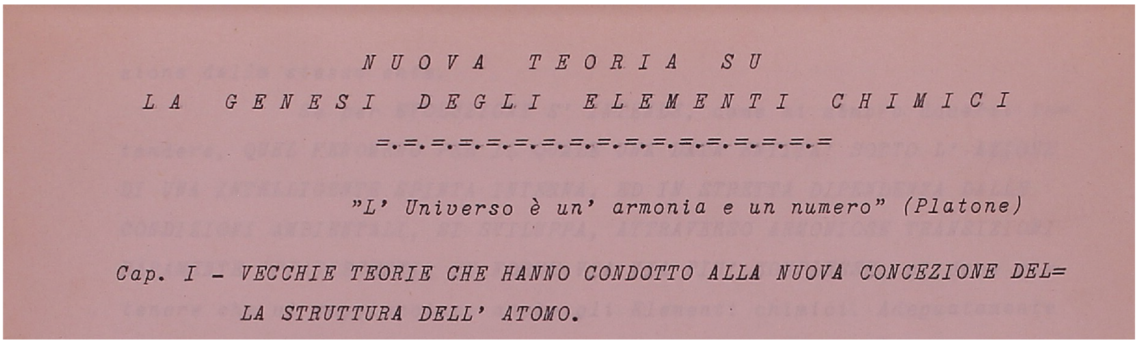
a) all the elements ought to originate from a single one, and;

b) all the elements are linked to one another trough common characteristics resembling genetic links in living organism;

It is unclear if Garosi is trying to mix ancient, pre-scientific, theories with modern ones, or borrowing their prestige to support his claims; drawing inspiration from old and new paradigms is nonetheless a feature that seems key to understand his work.

[5]Our translation; the original is "Il tutto è uno," Garosi, Nuova Teoria sulla Genesi degli Elementi Chimici, pg. 1

[6]"Se per EVOLUZIONE S'INTENDE, come mi sembra doversi intendere, QUEL FENOMENO PER IL QUALE UNA DATA ENTITA' SOTTO L'AZIONE DI UNA INTELLIGENTE SPINTA INTERNA, ED IN STRETTA DIPENDENZA DALLE CONDIZIONI AMBIENTALI, SI SVILUPPA, ATTRAVERSO ARMONIOSE TRANSIZIONE RARAMENTE APPARISCENTI, IN FORME VIA VIA PIU' COMPLESSE, si deve ritenere che vi soggiaciano anche gli Elementi chimici", Garosi, ibid.; all caps from the original work. In English (translation by the Authors): If by EVOLUTION we mean, as it seems we should, THAT PHENOMENON BY WHICH A GIVEN ENTITY, UNDER THE INFLUENCE OF AN INTELLIGENT INTERNAL IMPULSE AND IN CLOSE DEPENDENCE ON ENVIRONMENTAL CONDITIONS, DEVELOPS THROUGH HARMONIOUS TRANSITIONS THAT ARE RARELY CONSPICUOUS, INTO INCREASINGLY COMPLEX FORMS, then it must be considered that even chemical elements are subject to it.



N U O V A   T E O R I A   S U
L A   G E N E S I   D E G L I   E L E M E N T I   C H I M I C I
=.=.=.=.=.=.=.=.=.=.=.=.=.=.=.=.=.=.=

"L' Universo è un' armonia e un numero" (Platone)

Cap. I - VECCHIE TEORIE CHE HANNO CONDOTTO ALLA NUOVA CONCEZIONE DEL=
LA STRUTTURA DELL' ATOMO.

Figure 2: *Header of the first page of 1937 original typewritten edition of "The New Theory of the Origin of the Chemical Elements"; the quote is supposedly from Plato: "The universe is number and harmony", translation by the Authors.*

3.2 From Hydrogen to Uranium

As a reminder that a strong component of his theoretical work is actually rooted in experimental physics, Garosi begins with a review of the nuclear physics since the observations of Becquerel in 1896, mentioning the discoveries of Marie and Pierre Curie about radioactivity, to point out that helium nuclei, as byproduct of alpha decay, belong to the atomic nuclei of every chemical element heavier than hydrogen. His attention therefore focuses on the relation between the first two elements of the Periodic Table. He disagrees with the idea that helium is originated from the fusion of 4 hydrogen nuclei, and instead proposes a different interpretation, based on the hypothesis of the existence of three isotopes of hydrogen, each one heavier than the preceding of exactly one unit of atomic weight. The first twenty elements of the Periodic Table are analyzed in terms of atomic number and atomic weight: his first observation was of an alternating regularity between the elements. Leaps between them seemed to happen regularly in terms of 1 or 3 atomic weights: 3 from Hydrogen to Helium, 1 from Sodium to Manganese, then 3 from Manganese to Aluminium. With the exception of Beryllium, Nitrogen, and Argon, he devised a recurrent pattern between the first twenty elements:

H 1							HE 4
LI 7	BE (9)	B 11	C 12	N (14)	0 16	F 19	NE 20
NA 23	MG 24	AL 27	SI 28	P 31	S 32	CL 35	AR (40)
K 39	CA 40						

He notes two other patterns: *a)* the conservation of parity between atomic number and atomic weight, i.e. elements with even atomic number would have even atomic weight too, and *b)* the difference of 16 units in the atomic weights of homologous elements, for instance between Oxygen (16) and Sodium ($16 + 16 = 32$). All these findings helped him explain the deviations of Beryllium, Nitrogen, and Argon in terms of undiscovered isotopes: Beryllium-8, Nitrogen-13, and Argon-36. The elements on the Table then progress like:

$$\begin{array}{cccccc}
H^1_1 \rightarrow & He^2_4 \rightarrow & Li^3_7 \rightarrow & Be^4_8 \rightarrow & B^5_{11} \rightarrow & C^6_{12} \\
{\scriptstyle +H^1_3} & {\scriptstyle +H^1_3} & {\scriptstyle +H^1_2} & {\scriptstyle +H^1_1} & {\scriptstyle +H^1_2} & \\
 & & \rightarrow & Be^4_8 \rightarrow & B^5_{11} & \\
 & & {\scriptstyle +H^1_1} & {\scriptstyle +H^1_3} & &
\end{array}$$

Deuterium and Tritium, along with Hydrogen, take part in the formation of isotopes, as reconstructed in the graph above: $He^2_4 + H^1_3 \rightarrow Li^3_7$ originating the correct Lithium isotope in accordance with the rule of 1 or 3 steps between elements, while the two isotopes of Beryllium, Be^4_8 and Be^4_9, are generated by $Li^3_7 + H^1_1(hydrogen) \rightarrow Be^4_8$ and $Li^3_7 + H^1_2(deuterium) \rightarrow Be^4_9$.

While noticing that the rules of alternation of 1 and 3 steps between elements do not seem strictly enforced across the Table, he applies the other symmetries to Helium. Garosi therefore suggests the existence of five isotopes[7]:

$H + H \rightarrow He_2$

$H + D \rightarrow He_3$

$H + T$ or $D + D \rightarrow He_4$

$D + T \rightarrow He_5$

$T + T \rightarrow He_6$

We can see how Garosi is aware of using predictions as test bench of his theory, and this is fairly clear with Helium-6: the isotope itself has been discovered in 1936

[7]We will use Garosi's own notation, where H denotes Hydrogen, D as Deuteron, and T as Triton, the last two being the nuclei of each isotope; his notation for the isotope weight is also in use

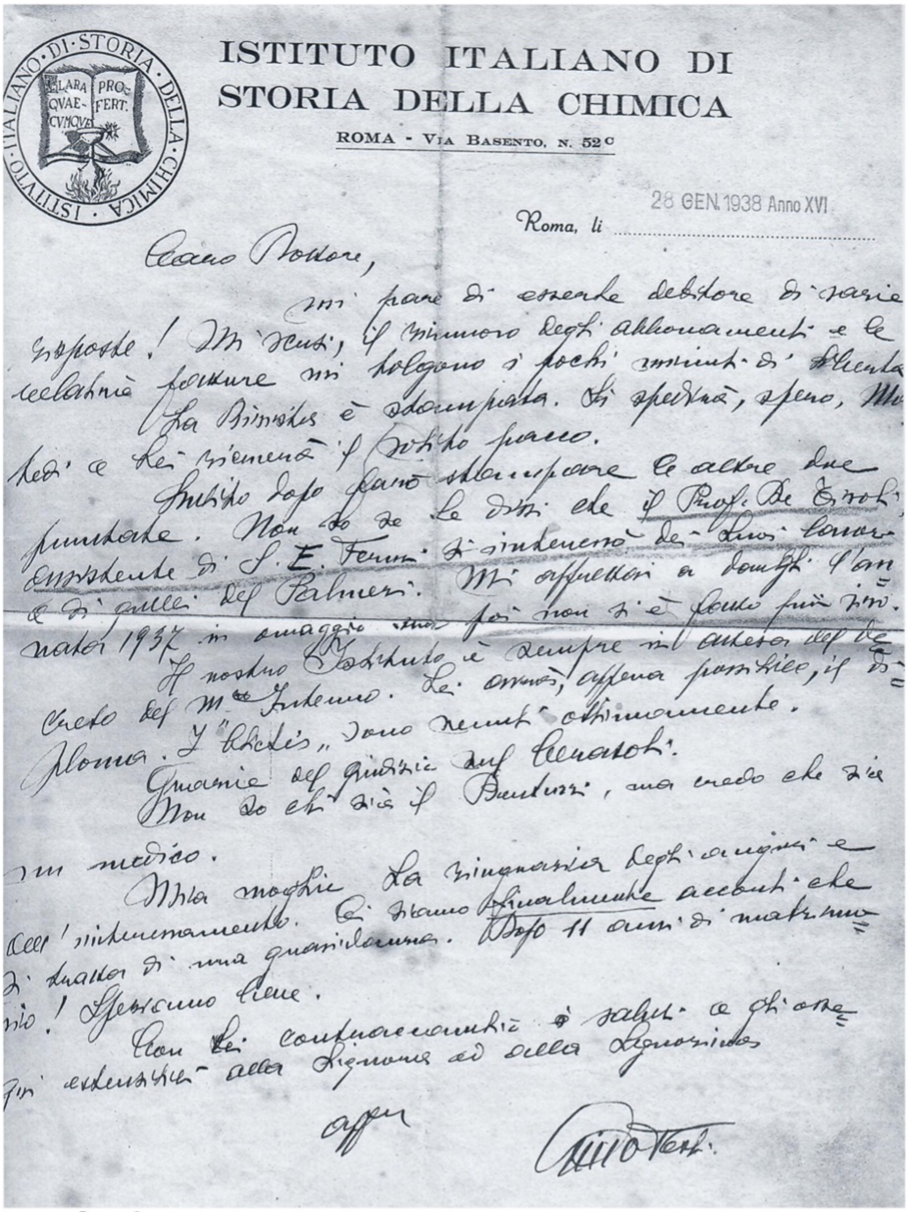
[25].

Garosi also proves to be fully aware of the developments in nuclear physics, reviewing its hypothesis to include the presence of *positron*, theorized by P. Dirac in 1928 and discovered in 1932 by C. D. Anderson: the proton is constituted by a neutron and a positron, together, to explain the presence of neutrons within the nucleus. Carlo Garosi is able to use his hypothesis to compute a more accurate value for the energy belonging to the process of Helium synthesis, testing, although indirectly, the consistency of his formulation. The hypothesis enables Garosi to redefine some basic concepts such as atomic weight, atomic number, isotope, incorporating the new experimental evidences - proving that he is well within the contemporary milieu - and ordering them, following general principles of simplicity, symmetry, logic. In doing so, Garosi masters correctly the mix of theoretical considerations and experimental evidences that defines the formulation of a scientific theory [11]. He is also able to draw predictions from that, postulating the existence of some isotopes and transuranic elements that will be indeed discovered in the following years [26].

4 Conclusion

Demonstrating a keen understanding of the evolving field, Garosi correctly recognized the 1934 experiment by Oliphant, Harteck, and Rutherford as the first evidence of tritium, a claim he made with conviction in 1936 [11]. Albeit limited, by political and geographical boundaries, Garosi successfully proved to be able to build a genuine scientific theory. It is also possible that his conclusions had some influence on the work of Enrico Fermi: in a private letter from 1938, Gino Testi[8] — one of the founders of the Istituto Italiano di Storia della Chimica and editor of the journal

[8]Gino Testi [Catanzaro 02/07/1892 – Rome 21/06/1951] graduated in pure chemistry and worked as a senior official at the Ministry of Finance until his retirement around 1948. He focused primarily on industrial chemistry and the history of chemistry, taught as a private lecturer at the University of Rome, and was among the founders of the *Italian Society for the History of Chemistry.* A World War I veteran, he also played a key role in the preservation of rare books and documents. Among his most successful works is the *Dizionario di alchimia e di chimica antiquaria* (1950). He contributed to various academic journals and was active in several academic societies.



ISTITUTO ITALIANO DI STORIA DELLA CHIMICA

ROMA - Via Basento, n. 52c

28 GEN. 1938 Anno XVI

Roma, li

Caro Dottore,

mi pare di essere debitore di varie risposte! Mi scusi; il [illegible] degli abbonamenti e le [illegible] forzate mi tolgono i pochi minuti di libertà.

La Rivista è stampata. Si spedirà, spero, martedì e Lei riceverà il [illegible] pacco.

Subito dopo farò stampare le altre due puntate. Non so se Le dissi che il Prof. De Tivoli, assistente di S.E. Fermi si interessò dei Suoi lavori e di quelli del Palmieri. Mi affrettai a donargli l'annata 1937 in omaggio ma poi non si è fatto più vivo.

Il nostro Istituto è sempre in attesa del decreto del M.ro Interno. Le manderò, appena possibile, il diploma.

I "Clichés" sono venuti ottimamente.

Grazie del giudizio sul [illegible].

Non so chi sia il [illegible], ma credo che sia un medico.

Mia moglie La ringrazia degli auguri e dell'interessamento. Ci siamo finalmente accorti che si tratta di una gravidanza. Dopo 11 anni di matrimonio! Speriamo bene.

Con Lei contraccambio i saluti e gli omaggi estensibili alla Signora ed alla Signorina.

aff.

Gino Testi.

Figure 3: *Private letter from Gino Testi to Carlo Garosi. From Garosi's personal archive, courtesy of Venera Dibilio Impallomeni.*

"Chimica "— informed Garosi that Aroldo De Tivoli[9] was interested in his research

[9] Aroldo De Tivoli [Buonconvento (Siena) 14/07/1888 – Florence 25/04/1972] served as assistant in the Central Office for the Preservation of the Standard Chorister at the *Regio Istituto Fisico (Royal Institute of Physics)* in Rome between 1922 and 1924. In 1924, he became Giulio Cesare

on the origin of the chemical elements and requested a copy of the 1937 manuscript. Professor De Tivoli was assistant director of the *Laboratorio Fisico della Sanità Pubblica*[10] (formerly *Ufficio del Radio*[11]), the official institution that provided financial and logistical support to Enrico Fermi's research team, the so-called Via Panisperna group[12]. Probably Garosi's theories circulated among Fermi's collaborators and may even have reached Fermi himself. The original letter is shown in Figure 3, while the English translation from the Authors is reported below:

> Dear Doctor [Garosi][13],
> It seems I owe you several replies! My apologies — renewing subscriptions and dealing with related invoices are depriving me of the few minutes of happiness I have. The [. . .] is printed. It will be shipped, I hope, on Tuesday, and you will receive the usual package.
>
> Right after that, I will have the other two episodes printed. I don't know if I told you that Prof. De Tivoli, one of the assistants of His Excellency Fermi, was interested in your work and that of Palmeri. I gave him the 1937 issue as a gift immediately, but then he never followed up.
>
> Our Institute is still waiting for the decree from the Ministry of Home Affairs. As soon as possible, you will receive the diploma. The [. . .] turned out excellently.
>
> Thank you for your opinion on Cerasoli.
> I don't know who Bentuzzi is, but I believe he is a doctor. My wife thanks you for your wishes and concern. We finally realized it is a preg-

---

Trabacchi's assistant at the *Laboratorio Fisico della Sanità Pubblica*, located at the Via Panisperna Institute. He taught mathematics for chemistry and natural sciences students in Rome (1928–1933), then he became lecturer in experimental physics (1935–1938) and mathematics (1938–1944). He also contributed several definitions to the *Enciclopedia Italiana*.

[10]"*Public Health Physics Laboratory*", translation by the Authors.

[11]"*Radium Office*" , translation by the Authors.

[12]See: `https://brunelleschi.imss.fi.it/nobel/iviapanisperna.html` and `https://media.accademiaxl.it/pubblicazioni/ScuolaFermi/st4.html#4`

[13]Authors' note

nancy — after 11 years of marriage! Let's hope everything goes well.
With warm regards, I extend my greetings to your wife and daughter.

Greetings

Gino Testi

Much more remains to be done, beginning with the personal archive of Carlo Garosi. Currently under the care of Venera Dibilio and Carlo Impallomeni, his direct relatives, the archive is an extraordinary opportunity to cast a light on an under-looked part of the Italian scientific community. The conspicuous amount of papers, letters, documents, magazines, and more, is a window on the stories of those local personalities and institutions that, far from the centres of the country, successfully established a proper research activity. This material needs to be indexed, organized and made fully available to others. The work of Carlo Garosi could become a case study for how the genesis and the acknowledgment of a scientific idea is far from a linear process.

Acknowledgments

The authors would like to thank Sandro Tatini, Tiziana Fantauzzo and Marzia Tatini for their help with the translation of the 1938 correspondence between Garosi and Testi.

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