

Positronium Chemistry: Origin, Development, and Historical Roots

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Received: Jun 11, 2025 **Revised:** Jun 28, 2025 **Just Accepted Online:** Jun 30, 2025 **Published:** Xxx

This article has been accepted for publication and undergone full peer review but has not been through the copyediting, typesetting, pagination and proofreading process, which may lead to differences between this version and the Version of Record.

Please cite this article as:

H. Kragh (2025) Positronium Chemistry: Origin, Development, and Historical Roots.

Substantia. *Just Accepted.* DOI: 10.36253/Substantia-3540

Abstract. Positronium science has for more than half a century been an important research area located somewhere between physics and chemistry. Based upon the study of positrons and the electron-positron atom called positronium, the field was originally part of atomic physics but soon became embraced also by the chemists. In this article I present a first preliminary study of the origin and early history of positronium science with an emphasis on its chemical aspects. It includes a so-called prehistory going back a long time before the discovery of the positron, at a time when the ether was sometimes conceived as consisting of pairs of oppositely charged electrons. Apart from positronium, the intertwined history of muonium – where the electrons are replaced by the heavier muons – is also described. More generally, the paper discusses from a historical perspective how various kinds of exotic atoms, antiatoms and superheavy atoms included, have become parts of the chemical sciences.

Keywords: positronium, muonium, exotic atoms, antimatter, hydrogen isotopes

1. INTRODUCTION

Terms such as ‘positronium’ and ‘muonium’ may not be commonly known, but for more than seventy years these exotic atomic systems or quasi-atoms have been studied intensely by a large number of researchers. They form today the basis of a flourishing interdisciplinary research area cultivated mostly by physicists and chemists but also by other scientists. In this essay I outline the development of the field from a historical perspective, paying particular attention to how positronium and like atomic systems migrated from theoretical physics to chemistry. Of course, it is not the only migration of this kind. On the contrary, since the mid-nineteenth century the chemical landscape has changed significantly as a result of new discoveries in physics.¹ Modern positronium chemistry, largely a child of quantum and nuclear chemistry, is as much physical in nature as chemical.

The standard history of positronium science – insofar there is such a history – is to trace the field back to either 1934, when electron-positron atoms were first hypothesized, or to 1945, when the term positronium was coined in the context of quantum physics. However, one can find earlier if only qualitative ideas of negative and positive electrons (not positrons) forming atom-like systems. These early and today forgotten ideas, summarized in section 2, may not belong to the history of positronium proper, but at least they belong to its prehistory. The alleged birth of positronium in 1934 is critically evaluated in section 3, to be followed by section 4 dealing with the first decade of the post-World War II development. At that time positronium was not yet recognized to be of interest to chemists, a situation that changed only in the 1960s with the childhood of positronium chemistry (section 5). In section 6, I take a broader and less historical look at positronium, muonium and related quasi-atoms (including antiatoms), briefly discussing their chemical nature and relevance.

Inevitably, the history presented here is selective and highly incomplete. But as I see it, even such a history is preferable to no history.

2. PREHISTORY: ELECTRONS AND ATOMS

Whereas positrons are positive electrons, in a historical context positive electrons (e^+) are not necessarily positrons ($\bar{e}$). In fact, the idea of light positive unit charges predates the discovery of the positron by more than half a century. Likewise, the idea of atomic systems composed of symmetrical combinations of positive and negative electrons can be found long before positronium was introduced.

As early as about 1870 the eminent German physicist Wilhelm Weber suggested that all matter and ether consisted of oppositely charged electrical particles of the same mass orbiting around each other.² By means of this hypothesis he thought that the chemical atom might in principle be explained. Without using the term 'electron' Weber's speculation was followed up by Karl-Friedrich Zöllner and a few other German physicists. However, it left no noticeable impact on the development that in the late 1890s led to J. J. Thomson's discovery of the (negative) electron. Even before Thomson's celebrated discovery Joseph Larmor proposed an electromagnetic theory based on the assumption of primordial particles which were "quantitatively alike, except that some have positive and others negative electrifications, the one set being ... simply perversions or optical images of the other set." He compared the two kinds of electrons – a term he used – with the chemists' optical isomers, stating that a simple atom or molecule might be "composed of a single positive or right-handed electron and a single negative or left-handed one revolving round each other."³

To the surprise of the physicists, the new electron appeared only as a negatively charged particle with no positive counterpart. And yet, although positive electrons as mirror particles of the negative ones failed to turn up in experiments, for a period of time they were considered as possible constituents of ether and matter.⁴

The British-Australian physicist William Sutherland worked on a variety of subjects, many of them on the borderline between physics and chemistry. Among

the research topics he investigated were Brownian motion, viscosity, spectroscopy, the ionic theory of solutions, the structure of water, and the intermolecular forces in liquids and solids.⁵ In a paper of 1899 he suggested that the free ether was made up of elementary particles, each of them composed of a positive and negative electron revolving around their centre of gravity.⁶ For the neutral ether particle, he coined the name 'neutron' which would later be adopted for a very different kind of elementary particle. "In free æther the positive and negative electron revolving ... round their centre of inertia form what I have proposed to call the neutron, the electric doublet," Sutherland stated two years later.⁷

Possibly without knowing of Sutherland's proposal, in his widely read textbook *Theoretische Chemie* Walther Nernst reintroduced the ethereal neutrons consisting of electron doublets. In agreement with Larmor and Sutherland, he wrote:

The relationship between positive and negative electrons evidently calls to mind the one between optical isomeric twins. It is a question of much importance whether a compound of the positive and negative electrons ($\oplus\ominus$ = neutron, an electrically neutral massless molecule) really exist; we shall assume that neutrons are everywhere present like the luminiferous ether, and may add that the space filled by those molecules is weightless, non-conducting, but electrically polarisable.⁸

To mention but one more example from the fin-de-siècle era, in 1901 young James Jeans examined in mathematical detail a hypothesis similar to the one of Sutherland and Nernst. Jeans considered "an atom as a collection of negative and positive ions [electrons], the negative ions each carrying a charge of electricity of amount $-e$, and the positive ions each carrying a charge $+e$."⁹ The only difference between the two species of electrons was the charge. Moreover, he stated that when a positive electron collided with a negative one, the two would annihilate according to the process



Thirty years later, Jeans' hypothetical process would reappear as Dirac's electron-positron annihilation. Without going into further details, it may not be too far-fetched to see in these early speculations an anticipation of what much later became known as positronium.

Still in 1906 atoms composed solely of positive and negative electrons was considered a possibility, if not perhaps the most likely one. Among several other atomic models, Oliver Lodge referred to the possibility that

... the atom may consist of a multitude of positive and negative electrons, interleaved, as it were, and holding themselves together in a cluster by their mutual attractions, either in a state of intricate orbital motion, or in some static configuration, kept permanent by appropriate connexions.¹⁰

Although the positive unit charge was soon identified as the hydrogen ion H^+ , eventually called the proton, the 'positive electron' continued to appear in the physics literature well into the 1920s if no longer as a mirror particle of the electron.¹¹

3. MOHOROVIČIĆ'S ELECTRON-POSITRON ELEMENTS

The modern positron was predicted by Paul Dirac in 1931 as the electron's antiparticle. In the same ground-breaking paper, he predicted the existence of antiprotons.¹² Two years later Carl Anderson reported his discovery in cosmic rays of positive electrons, which he called positrons. However, Anderson's discovery was unrelated to Dirac's theory and it was only later in 1933 that the positron was identified with the antielectron. As Dirac realized, this particle would quickly annihilate with a negative electron ($e^+ + e^- \rightarrow 2\gamma$), a process verified in 1934. When Dirac received the Nobel Prize in late 1933, the positron was widely accepted if not necessarily as an antielectron. With the discovery of artificial radioactivity in 1934, it was understood that positrons are not particularly rare or exotic. In some artificially produced nuclides, a proton transforms spontaneously into a neutron, a positron and a neutrino: $p \rightarrow n + e^+ + \nu$. Dirac ended his Nobel lecture with speculations

about antimatter composed of antiprotons and positrons. Perhaps, he suggested, half the stars in the universe were made of matter and the other half of antimatter.¹³

Dirac's speculation inspired an obscure Yugoslavian-Croatian physicist by the name Stjepan Mohorovičić to suggest a series of very light celestial atoms composed solely of electrons and positrons. The simplest of these exotic atoms, an electron and a positron revolving around their common centre of gravity, he called 'electrum'.¹⁴ Although Mohorovičić's paper in *Astronomische Nachrichten* was ignored for two decades, today it is widely recognized as the pioneering paper in positronium science. The first reference to it in a research publication dates from 1953 and presently it has received more than 200 citations.¹⁵

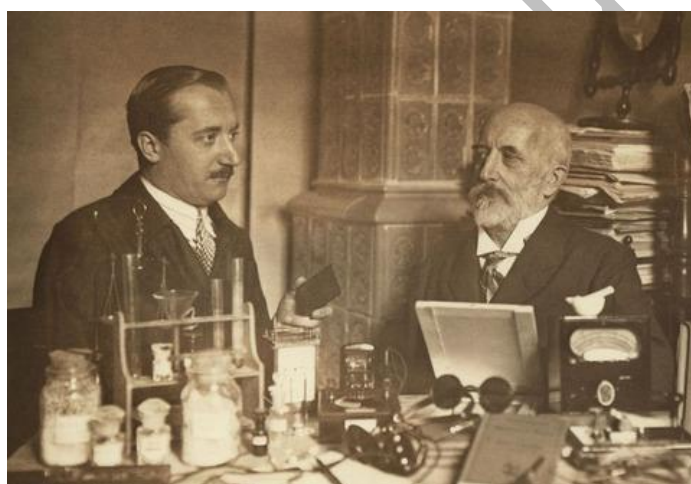


Figure 1. Stjepan Mohorovičić with his father Andrija Mohorovičić. Source: <https://cro2.salamander-studios.com/2019/06/03/stjepan-mohorovicic/>

The little-known Stjepan Mohorovičić was a productive and versatile but also decidedly unorthodox scientist. Born in Croatia in 1890, he was the son of the much better known Andrija Mohorovičić, the noted geophysicist and seismologist who in 1910 proposed the discontinuity between the Earth's crust and mantle named after him as the Moho discontinuity (Fig. 1). Mohorovičić junior published on a variety of subjects including geophysics, meteorology, fundamental physics, astronomy and philosophy.¹⁶ He was particularly interested in Einstein's theory of relativity which

he however rejected as philosophically unacceptable and lacking in experimental support.¹⁷ Like many other anti-relativists in the 1920s he maintained that the absolute ether was indispensable. There is little doubt that his poor reputation among mainstream physicists was a contributing reason why his 1934 paper on electron atoms attracted no interest at all.

Mohorovičić derived the theoretical spectrum of the electrum atom e^+e^- essentially by copying Bohr's treatment of the hydrogen atom from 1913, the only difference being that the heavy proton was now replaced with the much lighter positron ($M/m = 1836$). While in the case of the hydrogen atom the reduced mass

$$m^* = \frac{mM}{m + M} = \frac{m}{1 + m/M}$$

is close to the electron mass m , for electrum it is half this value, $m^* = m/2$. Likewise, while the ionisation energy of a hydrogen atom in its ground state is 13.6 eV, for electrum it is 6.8 eV. Mohorovičić considered electrum to be a new chemical element, if so far hypothetical, with atomic number $Z = 1$ and atomic weight $A = 0.0010863$ on the $H = 1$ scale. He proposed Ec as its chemical symbol. Because of electrum's large diffusion velocity, the element would have escaped the gravitational field of the Earth, but "the gas could easily be part of the corona of the fixed stars and the astrophysicists should at first look for it in the Sun's corona." To detect the element its characteristic line spectrum would suffice.

Electrum was not the only electron-positron atom considered by Mohorovičić (Fig. 2). He thought of a whole class of such elements, which he called "abarc" meaning non-heavy (from the Greek *barys* for heavy or weighty). For a few of them he suggested names and symbols, as for 'nobilium' (Nb) consisting of a nucleus of two electrons and four positrons ($Z = 2$) surrounded by two satellite electrons. The atomic weight of nobilium was thus 0.00436. Another of the abarc elements was 'slavium' with chemical symbol Sl and atomic number $Z = 3$ which he suggested might be identical to the old 'coronium' element hypothesized by earlier chemists

and astronomers. The possible existence of coronium was based on an unidentified solar spectral line which was only explained in 1939 as due to the highly ionised iron atom Fe^{13+} .¹⁸ By 1934, the hypothesis of coronium as a celestial element was still alive but no longer taken seriously in mainstream science.

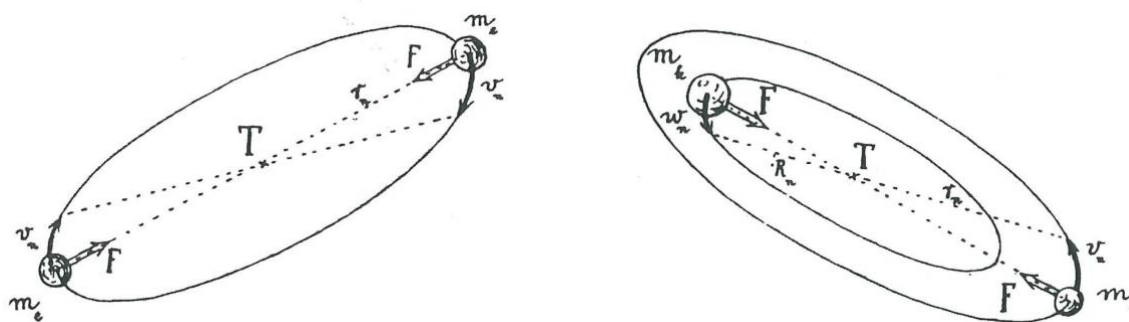


Figure 2. Mohorovičić's drawings in his 1934 paper of electrum (left) and nobelium (right).

Confident that the abaric electron-positron elements were real and would one day be detected, Mohorovičić ended his paper with an appeal to the chemists: "It will be up to the spectroscopists to search for these new elements, not only in the spectra of stars and nebulae but also in discharge tubes at high voltages. Should the predicted elements be confirmed in this way, they would also be of importance to pure chemistry."

Was Mohorovičić's electrum really the same as the later positronium? Did positronium science have its humble origin in his 1934 paper? Although the Croatian physicist clearly imagined an electron-positron atomic system, he did not identify the positron with Dirac's antielectron. For this reason alone, electrum was not just another name for the positronium system discovered many years later. Had Mohorovičić accepted the antielectron, which he knew of, he would have realized that the two constituents of electrum would rapidly annihilate. But his hypothetical hydrogen-like electrum atom was no less stable than the real hydrogen atom. To phrase it differently, in its spirit Mohorovičić's atom was closer to the fin-de-siècle ether speculations of Sutherland, Jeans and Nernst than it was to Dirac's ideas based

on relativistic quantum mechanics. On the other hand, there was the significant difference that while Sutherland and Jeans had ordinary atoms in mind, Mohorovičić thought of a series of entirely different atoms that might exist in parallel with the ordinary ones. I conclude that it is only half a truth to trace positronium history back to Mohorovičić's 1934 paper in *Astronomische Nachrichten*. The Croatian physicist is an interesting figure in the history of science but to credit him as "the father of positronium" is to go too far.¹⁹

4. FROM ELECTRUM TO POSITRONIUM

Other and perhaps better father figures may be American physicists Arthur Ruark and John Wheeler. In a short note in *Physical Review* dated 13 November 1945, Ruark suggested the name 'positronium' for the simplest electron-positron atom. "I think no physicist will doubt the existence of these unstable hydrogen-like atoms," he opined. As Ruark stated, possibly for priority reasons, he had contemplated the possibility of such an atom for several years: "In 1937 I conceived the idea that an unstable atom composed of a positron and a negative electron may exist in quantities for spectroscopic detection."²⁰ Shortly before he submitted his note, he had learned that Wheeler, by then a well-known nuclear physicist, had just submitted a longer and more detailed paper on the same topic. However, whereas Ruark's note appeared in print already on 1 December 1945, it took until 11 October 1946 before Wheeler's article on 'polyelectrons' appeared in the *Annals of the New York Academy of Sciences*.²¹

While both authors unknowingly reproduced some of Mohorovičić's results, such as the size and optical spectrum of the positronium atom, they went much beyond the Croatian physicist by recognizing that the positron was an antielectron and that the atom would therefore decay by annihilation into pure radiation energy. By taking into account the electron's spin – something Mohorovičić had ignored – they realized that the positronium system could exist in two states, one a singlet (para-positronium with antiparallel spins) and the other a triplet (ortho-positronium

with parallel spins). While para-positronium decays into two photons, in the case of the ortho form the result is $e^+ + e^- \rightarrow 3\gamma$. For the first state Wheeler calculated a lifetime of the order 10^{-10} s and for the second 10^{-6} s (the presently known values are 1.3×10^{-10} s and 1.4×10^{-7} s). Ruark and Wheeler both discussed various means by which the hypothetical positronium might be revealed experimentally but without advocating a definite experiment.

As to nomenclature, Wheeler spoke of 'polyelectron' generically and for the simplest systems he chose the names 'bi-electron' and 'tri-electron.' The associated symbols were P^{+-} and either P^{++-} or P^{+--} . The hypothetical positronium molecule Ps_2 was designated P^{++--} . Wheeler was the first to consider the question of whether a positron can attach itself to an atomic or molecular system and form compounds such as Cl^-e^+ and H^-e^+ . In addition to the suggestion of polyelectrons, Wheeler also considered the polarisation of photons from e^-e^+ annihilation. His remarks on this issue have later been interpreted as "the first clear and transparent written description of what quantum entanglement really is."²²



Figure 3. John Wheeler. Source: Niels Bohr Archive, Photo Collection.

In his autobiography from 1998, Wheeler recalled that for some time he had been fascinated by the idea that everything was built of negative and positive electrons:

I just explored some of the atoms and molecules that could be constructed from electrons and positrons alone, and calculated their properties. The simplest such atom ... is now called positronium, and its properties have since been extensively studied. It has the purity of my early dreaming. Unadulterated by quarks and anything else, its properties can be wholly understood in terms of the electron, the positron, and the photon. Later, I went further, calculating how a large collection of positronium atoms might behave. Liquid positronium should be superconducting.²³

The Ruark-Wheeler positronium was no less hypothetical than Mohorovičić's electrum, but within a few years it metamorphosed into a real entity. Born in Vienna in 1917 to a Jewish family, Martin Deutsch emigrated to the United States where he later became a physics professor at MIT.²⁴ In a series of experiments, he measured the delayed gamma photons emitted by positrons stopped in gases and gas mixtures. Correlating the e^+e^- annihilation rate to the gas pressure, he found in his data convincing evidence for a bound state of the two particles. As he concluded in a brief paper dated 13 March 1951, the data were "proof of the abundant formation of positronium."²⁵ It took another thirty years until the existence of the negative ion $e^+e^-e^-$ predicted by Wheeler in 1946 was demonstrated experimentally.²⁶

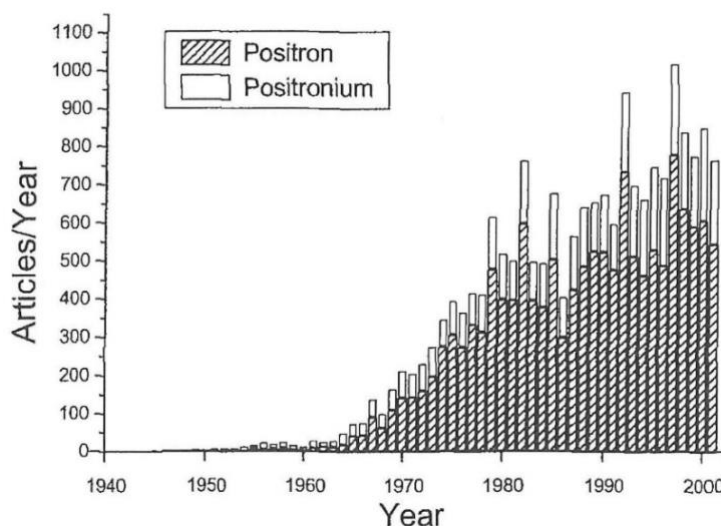


Figure 4. The growth of positron and positronium science. Reproduced from ref. 46, p. 8.

The confirmation that positronium really existed, albeit as an ephemeral atomic system only, was followed up by a large number of experimental investigations that placed positronium physics as a new and exciting branch of nuclear and particle physics. In July 1965 the First International Conference on Positron Annihilation was held in Detroit. As shown in Fig. 4, the number of research publications in the new field grew dramatically, in the early period dominated by workers in the USA and USSR. It was in this early period that positronium, conceived as an element, was designated the chemical symbol Ps first appearing informally in a 1959 paper in *Physical Review*.²⁷

Still by the mid-1950s positronium science was almost identical to positronium physics. It was a research area predominantly cultivated by particle physicists with the aim of understanding the fundamental interactions between radiation and matter. But a few physicists realized that since positronium was an atomic system, hence a kind of element, it might just as well belong to the chemical sciences. As two American physicists wrote in an extensive review article:

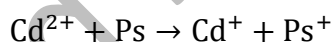
The formation and decomposition of positronium is related to the properties of the molecules of the surrounding gas; in a sense, it can be regarded as a chemical problem It would seem natural that the work should be continued by chemists, to whom

positronium should present the challenge of a new element whose chemical properties have to be classified; and, owing to its extreme simplicity, it may be an element of particular value for the understanding of the mechanism of chemical reactions and for the study of the nature of the chemical bond.²⁸

The article was reviewed in the *Journal of the American Chemical Society* the following year, with the reviewer, the chemist Richard W. Dodson, citing approvingly the above quotation.²⁹ This may have been the first time that the term positronium appeared in a high-ranking chemical research journal.

5. BEGINNINGS OF POSITRONIUM CHEMISTRY

Early workers in what came to be positronium chemistry were physicists or nuclear chemists who were primarily interested in how positrons and positronium interacted with matter in either the gaseous, liquid or solid state. For example, a study of 1959 looked at electrochemical reactions in aqueous solutions such as the reduction process³⁰



John Lee, a young Australian scientist, completed in 1960 a Pd.D. dissertation on “The Chemical Behaviour of Positronium” which he later developed into an influential monograph co-authored by James Green.³¹ Although the title of the book was *Positronium Chemistry*, it was not chemistry in the traditional sense accepted by the majority of working chemists at the time. Correspondingly, in this early phase almost all research on positronium chemistry was published in physics and not in chemistry journals.

The new field was thoroughly interdisciplinary and from the late 1960s onwards it was increasingly cultivated by scientists with a background in the chemical sciences. Articles on positronium began to appear in the *Journal of Physical Chemistry* and, more frequently, in the *Journal of Chemical Physics* established in 1932. One of the pioneers was the German-born Hans Ache, a professor of chemistry who

contributed with several papers in the mentioned journals. In an introduction of the subject to readers of *Angewandte Chemie—International Edition*, he described positronium as “the lightest isotope of hydrogen” and “an analog of the hydrogen atom ... in which the proton is replaced by a positron.”³² As he added, no doubt correctly, this atom-like system was “probably not so well known” to chemists and yet it was of great value with regard to a number of chemical problems. In a later review article based on a conference held in 1977, he commented on the short history of positronium chemistry:

The area of Ps chemistry was and still is made by scientists trained by physicists. However, despite all the pioneering efforts by these groups, chemists remained rather indifferent to this new atom and it was only during the past five to seven years that chemists of all persuasions have become more and more involved in the chemistry of this exotic atom ... We have successfully finished the first stage and ... we have to direct our attention now to the second stage, a task for which we need the cooperation of chemists in all areas of chemistry reaching from nuclear and radiation chemistry to bio- and enzyme chemistry.³³

Positron and positronium chemistry was not only cultivated by Western scientists but also attracted much interest in the Soviet Union, where the leading authority was Vitaly Goldanski, director of the Institute of Chemical Physics under the USSR Academy of Sciences. In 1968 he published a comprehensive book-length review of positron annihilation and positronium science which he described as “one of the most important branches of contemporary nuclear chemistry ... a tool for investigating the physical and chemical properties of matter.”³⁴ Goldanski was fascinated by the new light atoms, which apart from positronium also included the more recent muonium atom in which the positron is replaced by the 207 times heavier positive muon. He counted both of the new atoms as isotopes of hydrogen, suggesting that the centre of mass in positronium might count as a kind of immaterial nucleus.³⁵

Although the history of the muon (μ) is very different from that of the positron, muonium history has much in common with the older and better known positronium history.³⁶ The negative muon μ^- is conventionally called a muon and the positive muon an antimuon. The existence of an atom-like entity with an electron (e^-) revolving round a muon (μ^+) was proposed in a paper of 1957 in which the term 'muonium' was also introduced.³⁷ Three years later, the muonium particle was found experimentally by a team led by Vernon Hughes, a respected nuclear and particle physicist at Yale University. In a popular article in *American Scientist*, he described muonium as a new atom remarkably similar to the hydrogen atom except for its mass and short lifetime. Whereas the reduced mass m^* of positronium is $m/2$, for muonium it is close to that of the hydrogen atom. Correspondingly, the Bohr radius of muonium is 1.0043 times that of the hydrogen atom, and for the ionization energy the ratio is 0.9957. Hughes briefly suggested that the exotic atom might be of relevance to the chemists: "It seems clear that a substantial field of research in muonium chemistry may be available for study."³⁸ And indeed, within a few years 'muonium chemistry' evolved as a subfield in parallel with the larger and slightly older field of positronium chemistry.³⁹

Muonium looks in some respects like hydrogen, but there is the crucial difference that the first atom only survives for a period of approximately 10^{-6} s before the muon decays into an electron and two neutrinos. This happens to be of the same order as the lifetime of an ortho-positronium atom. But whereas the two components of positronium annihilate, in muonium the instability is caused by the muon's intrinsic lifetime of 2.2×10^{-6} s.

As IUPAC has accepted the chemical symbol Ps for positronium, so muonium has been assigned the symbol Mu. Whereas the first is a particle-antiparticle system, the latter is not. The muonium analogue of positronium would be the bound system ($\mu^+ \mu^-$), which is sometimes called 'true muonium.' Alas, this system is as yet hypothetical only. Since muons and electrons both belong to the lepton family of elementary particles, physicists have recently coined the term 'leptonium' as a

common name for positronium and muonium. However, it is not much used and 'leptonium chemistry' not at all. (While 'positronium' and 'muonium' are both accepted by the *Oxford English Dictionary*, 'leptonium' is not).

Paul Percival, a Canadian expert in muonium chemistry, reflected in a paper of 1990 on the nature of the new field.⁴⁰ Did it belong primarily to chemistry or to physics? As Percival pointed out, from a chemical point of view the properties of the muon are irrelevant as the chemistry is solely determined by the electrons. "Implicit in any definition of muonium chemistry is a demarcation of the boundary between chemistry and physics," he stated. Since no such intrinsic boundary seems to exist, he adopted a pragmatic criterion, namely to "define chemistry as what chemists do." According to Percival:

Muonium chemistry is defined by the activities of the handful of chemistry groups doing experiments at meson factories ... These few researchers cover an amazing breadth of science ... [and] they share expertise in radiation chemistry, molecular spectroscopy, chemical kinetics, molecular dynamics and more.

Before leaving muonium it is worth calling attention to the much heavier but still related atom called *muonic helium* first observed in 1980.⁴¹ This system consists of an electron revolving around an ordinary He-4 nucleus with a negative muon orbiting very close to it. The effective nucleus or pseudo-nucleus (α, μ^-) is thus of charge +1, namely an alpha particle combined with a negative muon. Despite its two protons, muonic helium behaves chemically more like hydrogen than helium. For this reason, it is sometimes considered a heavy isotope of hydrogen.

Positronium science was born in physics and subsequently migrated not only to chemistry and astronomy, but also to biology and medicine.⁴² Although the evanescent electron-positron atoms may seem to be strange creatures, today they are produced routinely and in large numbers also outside the laboratory. In positron emission tomography (PET), a medical technique dating from the 1960s, a radioactive positron-emitting tracer, such as ^{18}F , is injected into the patient. With a

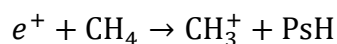
half-life of 110 min the fluorine isotope decays into ^{18}O and a positron which annihilates with an electron in the body. As a result, gamma photons are produced and these can be detected in the scanning device.⁴³ About 40% of the annihilation processes occurs through the formation of short-lived positronium atoms. With recently invented PET systems it has proved possible to study the photons from positronium annihilation and in this way to construct positronium images of the human brain.⁴⁴

One gets an impression of the rapid growth of positron and positronium science through a bibliography compiled in 1975 which includes references to 2,449 research articles published in the period 1930-1974. In the introduction to the bibliography, the compiler wrote: "It is apparent that positrons are finding increasing applications in physical chemical measurements, including chemical dynamics and mechanisms."⁴⁵

Half a century after Deutsch's discovery the annual output of papers related to positron annihilation and positronium chemistry, broadly conceived, was about 800 of which almost half were of a chemical nature.⁴⁶ By June 2025 the Web of Science records a total of 6,250 papers with 'positronium' as a topic and 3,147 papers with the term in its title. The corresponding numbers for 'muonium' are 2, 270 and 1,253, respectively. It should be noted that in the more recent literature the terms 'positronium' and 'muonium' often appear under their symbols Ps and Mu. To mention but one example, in a 2006 paper titled "Relativistic Ps^- and Ps " the term positronium appears 12 times and that of Ps 68 times.⁴⁷

The atoms of ordinary gaseous elements, like hydrogen, form molecules (H_2). As Wheeler briefly suggested in 1946, the same may be the case with positronium forming Ps_2 or P^{++--} in his notation. However, the theoretically possible Ps_2 molecule called dipositronium defied observation for a very long time. Only in 2007 did laboratory experiments provide unambiguous evidence that the molecule had been created and in this sense exists.⁴⁸ It also took a long time before the theoretically predicted positronium hydride (HPs) was turned into a reality. Consisting of one

proton, two electrons and one positron, the stability of the molecule was predicted in 1951. Forty-one years later a group of physicists at Aarhus University, Denmark, created it by bombarding methane with an intense beam of positrons:⁴⁹



As far as the natural occurrence of either free Ps or Ps₂ is concerned, all searches have been negative. As Mohorovičić pointed out back in 1934, it should be relatively easy to detect the entities by means of their emitted optical spectral lines. Later astrophysicists agreed that this was indeed a possibility,⁵⁰ but so far there is no generally accepted observation of the spectral signatures from celestial positronium atoms. Nonetheless, astrophysicists and astrochemists have studied in detail the numerous gamma photons emitted by positron-electron annihilation in the interstellar medium and by a variety of stars. As most of these photons are known to have their origin in Ps atoms, these atoms may be said to have been indirectly observed in cosmic space.

6. DIGRESSION ON ELEMENTS, MATTER AND ANTIMATTER

As mentioned, in the early literature positronium was sometimes described as a superlight isotope of hydrogen.⁵¹ However, other authors have argued that this is a misconception. According to David Walker, "Positronium (e^+e^-) is not a hydrogen isotope. In this species there is no nucleus because the two particles are of equal mass."⁵² Walker believed in a light isotope of hydrogen, but this isotope was muonium and not positronium. The majority of scientists undoubtedly agree, if from different arguments, that positronium does not qualify as a hydrogen isotope. Two isotopes have the same number of protons but a different number of neutrons, which is obviously not the case with positronium and hydrogen. Moreover, "isotopes always have similar chemical reactivities [whereas] Ps reacts with molecules very differently from H ... it is inappropriate to consider Ps a light isotope of H."⁵³

But then, what is an isotope? The current IUPAC definition states that two nuclides are isotopes if they have the same atomic number Z but different mass

numbers A . It does not refer explicitly to protons, neutrons or chemical properties. The view of muonium as a hydrogen isotope has been sanctioned by IUPAC which in 2001 recommended names for muonium and the three hydrogen isotopes protium, deuterium and tritium. Despite the absence of a proton, "Muonium ... can be regarded as a light isotope of hydrogen," we are told.⁵⁴

The question of isotopy is closely related to the question of whether or not positronium (or muonium) should occupy its own place in the periodic system. If muonium is admitted as a hydrogen isotope, like deuterium and tritium, it already has a place. But what about the non-isotopic positronium? One can simply declare it a non-element outside the periodic system, end of story. The three editors of *Principles and Applications of Positron and Positronium Chemistry* thought otherwise, namely that the element-like positronium deserved a place in the system. "It should have its own special place in the Periodic Table of the Elements, as the Period 0 (zero), and Group 1, or 1A."⁵⁵ This is how positronium is placed in a revised and unconventional periodic system, an element preceding hydrogen in group 1 and with an atomic weight of 0.0011. The proposal, the only one of its kind, was politely ignored.

Not only do positronium and muonium have chemical properties, it has also been argued that the same is the case with muonic helium and the μ^+ particle itself. According to two Iranian quantum chemists:

The positively charged muon is similar to the proton from the structural and bonding viewpoint and deserves to be placed in the same box of hydrogen in the Periodic Table ... The same reasoning may be applied to place muonic Helium, as a composite system, also in the box of hydrogen.⁵⁶

If the positive muon is admitted as a hydrogen isotope – which is not the standard view – it would be another isotope lighter than hydrogen (protium) if not quite as light as positronium.

Although positronium includes an antiparticle, it is not an antiatom in the ordinary sense of the term. The simplest antiatom is a system composed of a positron orbiting an antiproton ($\bar{H} = \bar{p}e^+$), which physicists at CERN succeeded to create in 1995. Antihelium with two antiprotons, two antineutrons and two satellite positrons has not been detected in either the laboratory or in nature, and nor have heavier antiatoms. Only a few simple antinuclei have been produced, namely (apart from the antiproton) two helium antinuclei $\bar{He}$ of mass numbers 3 and 4.⁵⁷ Nonetheless, atomic and even molecular antimatter is theoretically possible and has attracted much attention by physicists, astronomers and chemists (not to mention science fiction authors). An example from the literature is the massive volume *New Directions in Antimatter Chemistry and Physics* published in 2011.⁵⁸ The term ‘antimatter chemistry’ turned up in 1994, but it seems to be a hyperbole or misnomer insofar that the field is largely restricted to purely theoretical investigations or to experiments with antihydrogen.

If antimatter consisting of antielements exist, it will have the very same chemistry as ordinary matter. As the Swedish physicist Hannes Alfvén, an advocate of cosmic antimatter, said: “The only difference between water and antiwater is that a mixture of the two would generate tremendous energy.”⁵⁹ In an antiworld the antielements would be organized in a system completely symmetric to the well-known periodic system. A few people have speculated about such an antisystem as an extension of the ordinary one, for example by providing the antielements with negative atomic numbers ($Z = -1$ for $\bar{H}$, $Z = -2$ for $\bar{He}$, etc.). Unfortunately, the Wikipedia article on antimatter claims that “a complete periodic table of antimatter” was envisaged by Charles Janet, a French naturalist and amateur scientist, even before Dirac came up with the idea of antiparticles.⁶⁰ Perhaps needless to say, the claim is unfounded.

7. CONCLUSIONS

As shown in this paper, the electron-positron atomic system known as positronium has historical roots that can be traced back many years before the positron was discovered in 1933. The atom, as it is generally called, was hypothesized twice, first in 1934 by Mohorovičić and next in 1945-46 by Ruark and Wheeler. However, the positive charge was only identified as an antielectron in the latter case. As far as the actual discovery of positronium is concerned, priority belongs to Deutsch's work of 1951. Initially seen as a playground for theoretical physicists, since the late 1950s chemists increasingly studied the exotic and short-lived atom. The early development of positronium chemistry and its place in the scientific landscape is sketched in the present paper. The same is the case with its sister subfield called muonium chemistry.

Some of the general problems concerned with positronium and muonium refer to their status as chemical entities. They are atoms of a sort, but of what sort? If they are atoms, supposedly there are also corresponding elements and like all other elements these should occupy places in the periodic system. Chemists discussed these and related questions early on, with the result that today muonium is widely accepted as an unusual isotope of hydrogen. Despite having distinct chemical properties, positronium is not generally counted as an element. Some of the general questions relating to positronium and muonium are relevant also to other forms of exotic matter including antimatter, superheavy elements, and protonium ($p\bar{p} = p^+p^-$). These questions have been discussed in the literature from a philosophical point of view,⁶¹ but they are not part of the present paper.

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