



Scientific legacy of Robert L. Platzman. Preliminary report[☆]

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Abstract

One of the founders of radiation physics and chemistry, Platzman (1918–73) taught us elements of our current understanding such as the nature of the hydrated electron, the basic theory of the yield of ions and other initial-product species, and the importance of the oscillator-strength spectrum. In addition to all this seen in the literature, he left many unpublished materials, some of which contain stimulating thoughts and valuable ideas for experimental and theoretical work in the 21st century. © 2001 Published by Elsevier Science Ltd.

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1. Prologue

It was a great fortune for me to have been a collaborator with Robert L. Platzman in the 1960s. A portrait probably taken in this period is seen in Fig. 1. Over the past two years I have reviewed the contents of his office files. My purpose is to find materials suitable for an archive for possible studies by scholars in the future; this archive will be permanently stored in the Joseph Regenstein Library of The University of Chicago. The materials thus identified include unpublished manuscripts, notes, and correspondence with giants such as Bethe, Franck, and Herzberg. What follows is an initial and preliminary report on some of my findings.

2. Biographic sketch

Robert Leroy Platzman was born on 23 August 1918 in Minneapolis as the elder of two sons in a middle-class family. His father had an accounting position in a motion-picture company, and his mother was a teacher of piano.

Parenthetically, to see his life span against a perspective of the history of physical science, I point out notable scientists also born in 1918: Richard Feynman, Julian Schwinger, Jerome Karle (physical chemist, Nobel laureate in chemistry, 1985), Res Jost (Swiss mathematical physicist), and Abraham Pais (physicist and historian known for biographies of Einstein and Bohr).

Platzman received higher education at The University of Chicago, culminating at age 24 with a Ph.D. in chemistry for work (Platzman, 1942) directed by James Franck. Then, he joined in war efforts during the World War II, first on radar development at Massachusetts Institute of Technology and later on the Manhattan District Project at The University of Chicago. Specifically he belonged to a section of the project, led by Franck and aimed at studying physical, chemical, and biological effects of ionizing radiation.

In 1946–47 he studied with Niels Bohr at Copenhagen, and later traveled extensively in Europe (Platzman, 1948). This experience was decisive in forming the style of his work and life, characterized by deep thought, extensive knowledge in many academic disciplines, and affection for European culture. On return to the US, he began to teach students and to conduct research as a faculty member of Department of Physics at Purdue University. In 1958 he joined the staff of Argonne National Laboratory as Senior Physicist, where he stayed till 1965. During his appointment at Argonne,

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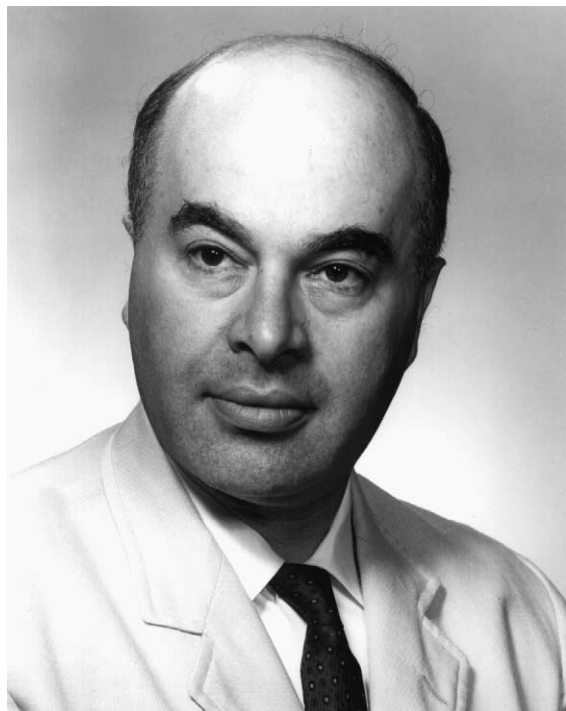


Fig. 1. A portrait of Robert L. Platzman, taken in about 1963.

he received a Fulbright Fellowship and Visiting Professorship at University of Paris, which enabled him to travel to Paris many times.

In 1965 he left Argonne to be a Professor of Physics and Chemistry of The University of Chicago, and also to assume the position of the Master of the Collegiate Physical Science Division. After carrying out duties at the University for nearly eight years, he was looking forward to a sabbatical to begin in the fall of 1973, when he was scheduled to be a Visiting Professor again at his beloved Paris. A heart attack deprived him of this prospect; he died on 2 July 1973.

A memorial service was held on 22 October 1973, where Fano and Hart (1974) presented tributes describing work of Platzman as physicist and chemist. A special issue of the *International Journal for Radiation Physics and Chemistry* (Vol. 7, No. 2/3, 1975) was dedicated to his memory, including a list of publications (on pp. 51–54), a tribute by Burton (1975), and an account of life and work by Haissinsky and Magat (1975).

He and his wife Eva (deceased in 1995) had three children: Loren, born in 1951, a computer software engineer; Elena, born in 1953, an international economist; and Kenneth, born in 1957, a computer specialist.

Robert's younger brother, George William Platzman, is also a scientist, specializing in fluid dynamics applied to the earth's atmosphere and oceans, Professor Emeritus, Department of Geophysical Sciences, The

University of Chicago. He made the file materials of Robert available for my study.

3. Major accomplishments, or the generally known legacy

One of the founding fathers of radiation physics and chemistry, Platzman is known for many outstanding accomplishments, as explained by Fano and Hart (1974), by Burton (1975), and by Haissinsky and Magat (1975). In addition, Fano (1975) wrote about the special style of work, and Inokuti (1975, 1985) gave further recollections of Platzman. What follows is a brief summary intended to provide a background for understanding some of the unpublished materials (to be discussed in Section 4).

3.1. Prediction of the hydrated electron

The most seminal of his accomplishments is the prediction (Platzman, 1953) of the presence of hydrated electrons in irradiated water, and more specifically, a proposal of verifying the prediction by observing a photoabsorption spectrum of the species. When he made this prediction, there were considerable objections, as seen in the Proceedings of the Highland Park conference, where Platzman (1953) presented his idea. However, by the late 1950s kinetic studies on the radiolysis of water led to the recognition of a reducing species other than the hydrogen atom, for which Platzman's hydrated electron must have been thought as a good candidate species. Almost a decade later, Hart and Boag (1962) unequivocally confirmed Platzman's idea by an innovative experimental feat, i.e., the first measurement of the absorption spectrum of hydrated electrons through the pulse-radiolysis method, which had been newly developed. Boag (1965, 1989) describes in detail the circumstances of this development. The key role of the hydrated electron in the radiation chemistry of aqueous solutions is now well established. An excellent historical review by Jonah (1995) contains a full account of studies on the hydrated electron.

Considerations on the hydrated electron were a natural extension of his thesis work (Platzman, 1942) on the spectra and photochemistry of negative ions in aqueous solutions. In this sense, it was fortunate that his extensive knowledge and experience found application in the radiation chemistry of water, a field of research new at that time. The topic of ions in aqueous solutions was further pursued, as seen in the two later papers (Platzman and Franck, 1952, 1954).

3.2. Subexcitation electrons

He innovated the notion of subexcitation electrons (Platzman, 1955), which are those electrons with kinetic

energies lower than the first electronic-excitation threshold in a non-metallic medium. They are abundantly produced under any ionizing radiation, lose kinetic energy gradually, i.e., only by exciting modes associated with nuclear motion; this occurs at a rate much less than the rate of energy loss by electronic excitation, which applies to electrons of higher energies. Therefore, subexcitation electrons are appropriate for consideration as a special entity. He first saw the need for the notion in the total ionization by alpha particles in helium admixed with various gases, observed by Jesse and Sadauskis (1955). The plateau values of the total ionization, observed at sufficiently high additive concentrations, were different for different additives, and were higher for additives of lower ionization thresholds; he interpreted the differences in terms of subexcitation electrons in helium.

For another example, the production of negative ions through dissociative electron attachment to molecules occurs mainly by subexcitation electrons. In general, a discussion of the primary processes of radiation actions would be incomplete at best without some consideration of subexcitation electrons. Recent years have seen extensive developments of this topic (Inokuti, 1990; Kimura et al., 1993).

3.3. *Theory of the yields of ionization and other initial species*

One of the fundamental problems of radiation physics and chemistry is to evaluate the number of ions, excited states, and other species that are produced as an immediate result of the absorption of ionizing radiation by any given medium. Platzman considered this problem in various contexts, including the yield of ionization in gases (Platzman, 1961). One of his innovations was an illuminating plot showing contributions of degrading electrons of different kinetic energies to the yield, as exemplified by Figs. 2 and 3 of Platzman (1961). We now call the plot the yield spectrum (Kimura et al., 1993).

He chose for the Ph.D. thesis work of his student, Miller (1956), a comprehensive study of ionization and excitation of helium gas, for which basic data such as electronic energy levels and electron-collision cross sections were better known than for other materials. Parenthetically, Miller later became famous as a computer scientist and high-level manager, culminating as Provost of Stanford University. Work on helium included several powerful ideas for the analysis of cross-section data, as explained by Fano (1975), and later led to a broader range of applications (Kimura et al., 1993).

3.4. *Oscillator-strength spectra*

By far the most decisive part of the cross-section data needed in the theory of the yields of initial species concerns glancing collisions of fast electrons or other charged particles, where electric-dipole interactions dominate according to the Bethe (1930) theory. Thus, the yields in a medium can be evaluated approximately if the dipole oscillator-strength spectrum, or the photoabsorption spectrum, of that medium is known over the complete range of excitation energies. More precisely, the yield of an initial species is approximately proportional to the dipole matrix element squared, viz., the oscillator-strength spectrum divided by the excitation energy, for the pertinent type of excitation. Platzman (1962b, 1967) called this idea “the optical approximation”.

In this sense, the study of the photoabsorption spectrum is of fundamental importance to radiation physics and chemistry. Note the proviso “over the complete range of excitation energies”, which is important not only because the energy transfer from charged particles is broadly distributed but also because the trustworthy establishment of the spectrum is possible by the use of sum rules, which requires the proviso. For this reason, Platzman pressed the need for measurements of photoabsorption and related spectra of materials of interest to radiation science, and campaigned for the use of synchrotron radiation for this purpose, as explained by Fano (1975).

3.5. *Superexcited states of molecules*

In examining early data on photoabsorption and other phenomena, Platzman (1960) displayed his characteristic logical rigor in pointing out that the transfer of energy in excess of the first ionization threshold of a molecule does not necessarily lead to ionization. He then stressed the need for elucidating properties of superexcited states (Platzman, 1962a, b), in which dissociation into neutral fragments and pre-ionization occur in competition.

To prevent a misunderstanding, the term “superexcited states” was coined by G.S. Hurst, and not by Platzman, as Platzman (1962b) states. He preferred the term “pre-ionization” (of a molecule) after Herzberg, to the term “auto-ionization”, which is now used far more frequently.

The urge of Platzman to study photoabsorption spectra and superexcited states of molecules has influenced many workers, and has resulted by now in an extensive bulk of data and a fair amount of knowledge, as summarized by Hatano (1994, 1999).

4. Unpublished materials

I found a number of unpublished manuscripts and notes with varying degrees of approach to completion. All the documents show the characteristics of Platzman's work, i.e., the high level of scholarship, the vast knowledge of physics and chemistry, the great care for details, and the clear expression of thoughts. Platzman was a perfectionist; in other words, he sent out a manuscript for publication only after he felt that he had done the very best possible. He had an exceptional talent for the use of languages, including French and German, and yet devoted tireless efforts for producing the best-possible expression of his ideas in both writing and speech. Some of the materials in his files indeed show that he edited his manuscripts repeatedly until he was fully satisfied, and even reprints of his published papers, perhaps in the anticipation of revised publications. Therefore, it is with considerable hesitation that I publicly discuss the contents of the unpublished materials; he would vehemently disapprove the present act of mine. After deliberations I see now two reasons to defend it.

First, it has been nearly three decades since his death. Thus, he has become in effect a part of history, not personally known to a majority of current radiation scientists. His thought process deserves studies by future scholars. Second, the unpublished materials contain matters of continuing interest, amply showing both breadth and depth of his thoughts and work beyond his publications. Some of the documents listed below contain substantial ideas of current significance, at least in my view.

The titles of the unpublished manuscripts and notes include the following, expressed in his own words here indicated by quotation marks. The order of the documents listed is chosen merely for convenience of the present discussion, and has no deeper significance.

“On the Moderation and Capture of Slow Electrons in Liquid Water”

An incomplete manuscript of 6 pages, written no later than 1957 (judged by the “Purdue University” address on byline) was probably intended to describe fully the hydrated electron and its spectrum. It probably corresponds to the paper mentioned by Boag (1989), who says, “In 1953 Platzman even had a paper in draft giving details of his calculations but this was never published”.

Origin of the Molecular Yield in the Radiolysis of Water by Gamma Rays

An incomplete manuscript of 5 pages seems to be an extended account of work reported at the Harrogate meeting (Platzman, 1962c).

“Fundamental Distinguishing Characteristics of the Biological and Chemical Action of Ionizing Radiation”

This manuscript of 22 pages is based on a talk given at the Conference on Certain Fundamental Aspects of Biophysical Science, held at Bethesda, Maryland on 15 January 1958. It is almost complete, reads nearly as well as his published papers, and contains ideas of present-day significance. Reading aloud the opening paragraphs, which are reproduced below, I almost hear Platzman's voice.

“In little more than half-century, radiobiology has grown to be a major branch of biology, at least as measured by the extent of research effort. Part of current interest stems, of course, from pressing practical questions, such as implications in the field of public health of relentlessly rising radiation levels in food, air, and other material; problems of radiation accidents, a constant increase in which also seems inevitable; and medical effects (with their concomitant political overtones) of fission and fusion weapons, both present, in weapons testing, and future, in possible warfare.

The great and widespread activity in the field also devolves, in part, upon the circumstance that it is almost uniquely easy to carry out a new, and by present standards, publishable experiment in radiobiology: the effect of () radiation on the () property of the () system under () conditions. This ease is a commanding attraction and to it much of the aimlessness of the literature must doubtlessly be attributed.

But the paramount goal of radiobiology must be, of course, not the elucidation of radiation effects as such, but the exploitation of radiation to advance understanding of the familiar and fundamental biological phenomena. In attempting to direct efforts toward this goal it should be clearly helpful to characterize those features of the action of ionizing radiation which distinguish it from other modes of altering the behavior of biological systems. There are three such features, at the primary or physico-chemical level: track effects, formation of molecular states of very great electronic excitation energy, and the influence of the dielectric dispersion of the medium on its reaction to initial products. They will now be surveyed briefly, with particular emphasis on aspects which are presently either unknown or ill-understood...”

The passage of the second paragraph including the four blanks contains a satirical epigram. The blanks within the parentheses in the manuscript were left by Platzman, to be filled by the reader with any descriptors

to generate titles of any number of papers. A similar remark may apply to radiation chemistry, too.

Of the three “fundamental distinguishing characteristics discussed by Platzman, only the “track effects” are a part of general knowledge among current radiation scientists. The “formation of molecular states of very great electronic energy” is considered by some of them, and “the influence of dielectric dispersion” is appreciated by a few of them.

“The Role of Dielectric Dispersion in the Action of Ionizing Radiation”

An almost complete manuscript of 17 pages, dated 14 April 1955, is substantial, and is significant even now. It presents a general discussion of the possible influence of the dielectric dispersion of a medium resulting from irradiation on the physics and chemistry leading to radiation effects. Examples include liquid water and proteins. Work on this topic does not seem to have been pursued as intensely as its importance warrants.

“Remarks on the Concept of ‘Density of Ionization’ in Radiation Chemistry and Biology”

An unfinished manuscript of 6 pages discusses the difficulties of evaluating the energy delivered per unit volume of an irradiated material, and points out errors found in the literature.

“The Final Fate of Electrons”

An unfinished manuscript of 8 typewritten pages, 11 handwritten pages and notes, and figure drafts was prepared on the basis of a talk given at the Fourth Congress of Radiation Research, held at Evian, France on 28 June through 4 July 1970 (which I did not attend). The typewritten part is “I. Introduction”. The handwritten part contains “II. Formation of Electrons”, “III. Interaction of the Electrons and the Medium”, “IV. Moderation of Electrons”, and “V. Chemical Action of Electrons”, the last three sections being notes rather than full texts. Nevertheless, this manuscript is of great interest because it deals with a vast topical area, and because it was produced late in his life.

Fig. 2 reproduces the first page of the handwritten part. It illustrates the beauty of his handwriting, and presents a refreshing charm in the present day, when most of us compose on a word processor.

“A New Type of Photochemical Reaction of Complex Negative Ions”

A preliminary manuscript of 4 pages with P. Pringsheim discusses polyatomic negative ions in aqueous solutions. It focuses on those ions which “exhibit a wide variety of absorption spectra and photochemical reactions which

depend upon the special constitution, rather than the negative ion character.

“On the α -Excited Fluorescence of Sodium Salicylate in Water”

A preliminary manuscript of 9 pages with A. Weinreb describes an estimate of the effects of subexcitation electrons (Platzman, 1962a) on the quantum yield of fluorescence.

Quantitative Evaluation of the Dispersion and Its Relation to Other Properties for Gases. I. Helium

A virtually complete manuscript of 30 pages with W.F. Miller is based on the Ph.D. thesis of Miller (1956), probably written in 1957. Through a thorough analysis of various data with the use of sum rules and other theoretical constraints, a complete spectrum of the dipole oscillator strength of He was determined. A highlight is the result that the oscillator strength of the strongest line, i. e., for the 2^1P excitation, is about 0.277, as announced in a related published paper (Miller and Platzman, 1957). This value agrees closely with 0.2762, the result of a highly reliable calculation by Schiff and Pekeris (1964).

This topic has been pursued by many workers, perhaps most recently by Berkowitz (1997) and by Yan et al. (1998). Compared with these recent studies, the work of Miller and Platzman looks remarkably good.

“On the Energy Dependence of Cross-Sections for Inelastic Collisions of Electrons with Atoms and Molecules”

An incomplete manuscript of 6 pages with W.F. Miller, also based on the thesis of Miller (1956), explains the power of a plot of data recommended by Fano (1954), which is now known as the Fano plot (Inokuti, 1971).

“On the Efficiency of Ionization by Positive Ions of Intermediate Velocity”

A nearly complete manuscript of 5 pages explains reasons why the yield of total ionization in a gas differs for different particles such as protons and alpha particles at the same low speeds. It seems to supplement a part of the discussion in Platzman (1961).

“A Theory of the Solar Corona”

A complete manuscript of 7 pages, dated 2 August 1948, discusses the origins of spectral lines of the solar corona, considering many aspects ranging from atomic ions, nuclei, and neutrinos. This document, together with the next two items, shows the remarkable breadth of Platzman’s interest and knowledge.

“The Perturbation of the Electronic System of an Atom by Radioactivity of Its Nucleus”

II. FORMATION OF ELECTRONS

It is a characteristic of matter, whether GASEOUS or CONDENSED, that an energy gap E_1 exists between the lowest possible state (the GROUND STATE) and the next-to-lowest, the FIRST EXCITED STATE. This is a basic result of the QUANTUM THEORY and is true for all forms of matter, excepting only METALS. Typical values of E_1 are in the range from about 4 eV to 10 eV, although there are a few special cases as high as 20 eV and as low as about 1 eV.

Electrons produced in any way within matter with $T > E_1$ lose kinetic energy very quickly in collisions which result in EXCITATION or IONIZATION — i.e., in forming PRIMARY PRODUCTS. The rate of ENERGY LOSS is of magnitude 10^{17} eV/sec, or GREATER, and almost nothing is competitive with this mode of retardation. (We shall disregard such complications as energy loss by bremsstrahlung, the behavior in the neighborhood of $T = E_1$, and the influence of LOW-LYING TRIPLET STATES, since they are readily treated and are irrelevant to our general considerations.)

Once $T < E_1$, the electrons lose energy much more slowly: they have a long lifetime and therefore greater ~~UNUSUAL~~ POTENTIAL for CHEMICAL INTERACTIONS. Numerous NEW PHYSICAL INTERACTIONS also enter the picture. These are, of course, the SUBEXCITATION ELECTRONS, and they play an essential role in radiation action on ALL FORMS of MATTER — again excepting metals.

The FINAL FATE OF ELECTRONS is necessarily a question of the behavior of subexcitation electrons.

Fig. 2. A sample of handwriting, from an unfinished manuscript entitled "The Final Fate of Electrons," based on a talk presented at the Fourth Congress of Radiation Research, held at Evian, France on 28 June through 4 July 1970.

“The Influence of Neutrino Absorption on Isotopic Abundance”.

These two items are notes on parts of the foregoing manuscript.

“Attempt to Study Excited States of H^- ”

This item is a set of extensive notes and calculations. Platzman carried out extensive calculations using the Hylleraas variational method on the possibility of an excited state of the hydrogen negative ion. His result was negative, in agreement with general knowledge of the present day. This material shows his interest and expertise in computational work, which may not be apparent in his publications.

“On the Possible Collective Excitation of Electrons in Atoms and Molecules”

An unfinished manuscript of 10 pages, written during the sojourn in 1959–60 at Paris, was meant to follow up the published paper (Platzman, 1960). It discusses the possibility of states resulting from “a simultaneous excitation of several or many electrons” in an atom or molecule that might account for “the major portion of the total oscillator-strength spectrum” at excitation energies of 15–25 eV. It explores possible reasons for the stability of such states. This item is accompanied by extensive correspondence with Fano, Lindhard, and Brandt.

Unlike other items, this manuscript was later given up. In a conversation with me in about 1963, Platzman basically discounted the above possibility, in view of the general success of approaches starting with a single-electron approximation. (Fano and Cooper, 1968)

Other materials in the office files include notes of lectures by Platzman and by others, and correspondence. He was also kind and generous to help other scientists who showed him manuscripts; some of his correspondence shows his extensive queries and suggestions on manuscripts by others on topics he must have regarded important. I am certainly a beneficiary of his great education in this respect.

5. Epilogue

The main point I learn from Platzman is the power of disciplined thinking backed up by a broad perspective and knowledge. My study of the Platzman materials remains incomplete now, and I intend to work further toward a full report in the near future. In the meanwhile I would welcome receiving queries or suggestions from anyone who shares interest in the subject matter.

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