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WILLIAM DRAPER HARKINS

*1873—1951*

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*A Biographical Memoir by*  
ROBERT S. MULLIKEN

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*Biographical Memoir*

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W. D. Perkins.

# WILLIAM DRAPER HARKINS

*December 28, 1873–March 7, 1951*

BY ROBERT S. MULLIKEN

WILLIAM DRAPER HARKINS was a remarkable man. Although he was rather late in beginning his career as professor of physical chemistry, his success was outstanding. He was a leader in nuclear physics at a time when American physicists were paying no attention to nuclei. Besides this, his work in chemistry covers a broad range of physical chemistry, with especial emphasis on surface phenomena. He was a meticulous and resourceful experimenter, as well as an enterprising one, who did not hesitate to enter new fields and use new techniques. He participated broadly, not only in the development of pure science but also in its industrial applications.

Harkins was born December 28, 1873, the son of Nelson Goodrich Harkins and Sarah Eliza (Draper) Harkins, in Titusville, Pennsylvania, then the heart of the new, booming oil industry. At the age of seven, he invested his entire capital of \$12 in an oil well that his father had drilled in the Bradford, Pennsylvania, fields. This investment returned his capital several times. Fortunately for science, the returns were not great enough to attract him permanently into oil production.

In 1892, at the age of nineteen, Harkins went to Escondido, California, near San Diego, to study Greek at the Escondido Seminary, a branch of the University of Southern California. The courses of study are described in the University of Southern

California Year-Book for 1891–1892, where Harkins is listed as a student. Apparently, the seminary (now the Escondido High School) served as a preparatory school for the university. Harkins attended the seminary for one year, but had to learn Greek elsewhere because it was discontinued the year he came; he enrolled in a general arts course. It seems that Harkins spent a few more years in Escondido, and made many friends, but there is no other information as to his activities while there.

Harkins entered Stanford University in 1896 at age twenty-three and received an A.B. in chemistry in 1900, at twenty-six. In 1898–1900, he was assistant, then instructor, in chemistry at Stanford. For the next twelve years, Harkins was professor and head of the Department of Chemistry of the University of Montana at Missoula, but he spent a considerable amount of time in postgraduate and postdoctoral work elsewhere. He did postgraduate work at the University of Chicago in 1901 and 1904, and at Stanford University, 1905–1906, culminating in a Ph.D. in chemistry from Stanford on June 10, 1908. He did research in Germany in 1909 and was at the Massachusetts Institute of Technology as research associate in 1909–1910. Harkins left Missoula in 1912 at age thirty-nine for an appointment at the University of Chicago, where he conducted further research the remaining thirty-nine years of his life.

In Missoula, Harkins took part in the life of the city and state: He was President of the Missoula City Board of Health from 1906 to 1912. He was chemist in charge of smelter investigations for the Anaconda Farmers Association (1902–1910), the Montana Copper Company of California (1904), and the U.S. Department of Justice (1910–1912). His first four scientific papers, published during the period 1907–1910, are devoted to arsenical poisoning of animals by smelter smoke, and related matters. In 1911 Harkins did some research for the Carnegie Institution of Washington. On June 9, 1904, he married Anna Louis Hathaway, who was head of the Department of English

at the University of Montana. She had also been a graduate student at the University of Chicago.

At the University of Chicago, with the opportunity to conduct pure research and to work with graduate students, Harkins' advancement was rapid. He was first assistant professor of general chemistry (1912–1914), then associate professor (1914–1917), then professor of physical chemistry. In 1916–1917 he was a professorial lecturer at the Mellon Institute for Industrial Research, and he lectured at the University of Illinois (1918–1919). In 1935 he was appointed Andrew MacLeish Distinguished Service Professor at Chicago. He was George Fisher Baker Lecturer at Cornell University in 1936–1937. In 1939 he retired officially at Chicago, but continued research with undiminished vigor until his death in 1951.

During World War I, early in 1915, Harkins began work on explosives for the Allies. Later in the war, he did special work for the army and the Chemical Warfare Service. Throughout his career, while carrying on notable work in pure science, he also contributed to applied science: as a consulting chemist with the U.S. Bureau of Mines, 1920–1922; consulting engineer, U.S. Air Service, 1924–1927; and consulting chemist, Chemical Warfare Service from 1927, Libby–Owens–Ford Glass Company from 1929, Universal Oil Products Company, 1930–1951, and United States Rubber Company, 1939–1941. During World War II, he was a member of the National Defense Research Committee (1941–1945). Civically, he also participated as a member of the Chicago Commission on Ventilation (1916–1928).

Harkins was also active in the affairs of the American Chemical Society. He was editor of the section of General and Physical Chemistry of *Chemical Abstracts* (1939–1951), chairman of the Chicago Section (1915–1916), chairman of the Division of Physical and Inorganic Chemistry (1919–1920), and councillor-at-large for a time. He was the recipient of the Willard Gibbs

Gold Medal of the American Chemical Society on May 28, 1928, in recognition of his work in surface chemistry and on nuclear structure and isotopes. He was also a vice president (chemistry) of the American Association for the Advancement of Science. Harkins was elected a member of the National Academy of Sciences in 1921, and at the annual meetings in Washington in April he took a lively part in the discussions. He was also a member of the American Philosophical Society, to which he was elected in 1925.

In Chicago, Harkins always lived with his family near the university (at 5437 Ellis Avenue). He and his wife had two children, Henry Nelson Harkins and Alice Marion Harkins. Henry Harkins (born in Missoula in 1905) obtained B.S. and M.S. degrees in physical chemistry, a Ph.D. in medicine (1928), and an M.D. in 1931, all at Chicago. He went on to a distinguished career in surgery.\* His M.S. thesis on surface tension of blood serum was completed under his father's direction in 1926. Marion Harkins won success as a concert singer. The members of the family were devout Episcopalians. The Harkins family had a summer home on Lake Michigan, at Lakeside, across the lake from Chicago. An active mountain climber in his youth in California and Montana, Harkins visited the Rockies annually for many years. At the time of his death, he had been paying daily visits to the hospital after Mrs. Harkins had suffered a stroke.

Harkins' contributions to pure science covered a wide spectrum in the field of physical chemistry, extending also into physics. When I came to Chicago as a graduate student in 1918, it was because I had read about Harkins' pioneering work toward the understanding of nuclear structure, a subject ignored at that time by American physicists. In fact, during the period 1913-1928, Harkins and his students were the only

\* G. Egloff, "Fathers and Sons in Chemistry," *Chemical and Engineering News* 22(1944):804.

Americans engaged in work relating to the structure of the atomic nucleus.

A perusal of the bibliography of Harkins' papers gives a perspective of his scientific interests and of his graduate students and other collaborators. In 1915 the diversity of his interests is already evident. His most extensive work was in surface chemistry (115 papers) and in nuclear and atomic structure and isotope separation (nearly 80 papers).

On the occasion when he received the Willard Gibbs medal, Harkins gave an address that shows something of his personality and the beginnings of his activity in his major fields of research. Following is a quotation from the introductory part of his talk.

"As an undergraduate, research appealed to me as one of life's greatest adventures, and I was attracted both to the very large, in astronomy, and to the extremely minute, in chemistry and physics. While the study of the atom and of radioactivity, then a new subject, had an extreme fascination, there were two subjects of investigation in physical chemistry which seemed to me of such minor importance that I took a firm resolution never to be enticed into working on either of them.

"These two fields of work were surface tension and solubility. To illustrate, let us consider surface tension. I did not realize that the importance of the study of surfaces and surface energy arises from the fact that the surface lies outside every body, particle or cell. To get inside from outside or outside from inside, the surface must be traversed.

"In 1909 I went to Germany to study with Fritz Haber, the chemist whose work on the synthesis of ammonia lengthened the World War by one or two years. On the first day of my stay in Karlsruhe, he invited me to lunch with him and his assistant at the leading hotel of the city.

"Haber insisted that as a visiting professor—I was then professor of chemistry at the University of Montana—only a prob-

lem of extreme importance should be given to me. He and his assistant rose, drank my health, and Haber said, 'He shall work on surface tension.' Unfortunately, or fortunately, I knew hardly enough German to object, and when much later I found that many of the world's greatest scientists had been interested in surface phenomena, I was thankful for this lack of knowledge."

The work revealed to Harkins the fascinating problems of surface chemistry and initiated his highly original work in that field. When he was able after his establishment in Chicago to resume that work, he began his investigation of the orientation of molecules in surfaces. He was one of the three (the others were W. G. Hardy and Irving Langmuir) who independently suggested the theory of orientation of molecules in surfaces. At Chicago in the winter quarter of 1913-1914, Harkins gave the earliest series of lectures on the theory of this subject. Thus began a long chain of steps, from improved experiments to improved theory to new experiments, which characterized Harkins' work in surface chemistry and related fields for forty years. The sequence was particularly fruitful because Harkins combined meticulous and ingenious experimental techniques with a knack for original interpretation of data. Now continuing the quotation from Harkins,

"After the completion of the experimental work, I returned to America in order to work on physical chemistry with A. A. Noyes and G. N. Lewis, both of whom have been awarded the Gibbs Medal. Here I met my second aversion, for A. A. Noyes stated that, under the grant from the Carnegie Institution which supported the work, it was expected that the general subject of research should be the theory of solutions, but the special subject solubility."

In the last year of his life, Harkins completed a book, published in 1952, *Physical Chemistry of Surface Films*, summarizing his work on the subject. The book contains an introduction



by Thomas F. Young, a younger colleague and a great admirer of Harkins. This introduction contains a paragraph that comments interestingly on the fruits of Harkins' work at M.I.T.:

"During the brief period which Dr. Harkins spent at the Massachusetts Institute of Technology in 1909-10, Professor A. A. Noyes was greatly interested in theories of solutions, and inspired an outstanding group of young men to investigate the subject. A remarkable series of papers came from the laboratory describing work done by or under the direction of A. A. Noyes, G. N. Lewis, W. C. Bray, W. D. Harkins, and others. Of course Harkins did not know then how important that work on the thermodynamics of electrolytic solutions would be to his own later investigations of surface phenomena, especially his studies of adsorption. In 1911 he published three papers presenting his researches on solubility carried out at the Massachusetts Institute of Technology. In later years he contributed about ten more papers on ionic interactions. The work of A. A. Noyes and his group aided G. N. Lewis in his discovery of the ionic strength principle. The latter once remarked that the principle was obtained within a few hours after he had picked up notes of a conference held some ten years earlier with Harkins."

Continuing further with the quotation from Harkins' Gibbs Medal address, first about his work on surface chemistry, "Now the greatest of solubility rules is 'similia similibus solvuntur' or 'like dissolves like.' This rule suggested that the experiments on surface tension might have given results more in accord with the theory if more complicated molecules, such as those present in the muscles, had been used. It is advisable, however, in scientific work, to use as simple materials as will give the desired behavior, so substances like butyric acid were considered. . . . a molecule of this substance possesses the interesting characteristic that at one end it is like oil, and at the other like water.

"Thus we may place a thick layer of oil on water and add butyric acid. The water-like ends of the molecules should be soluble in water, and the oil-like ends in the oil, but only at the interface between the two can both ends of the molecule be satisfied at the same time. From this point of view the butyric acid should be very much more soluble at the interface than in either oil or water, which is true. Furthermore, at the interface there should be a certain structure, since the molecules of butyric acid should, in general, be oriented with oil-like ends toward the oil, and water-like ends toward the water. . . .

"A later careful search in the literature showed that Hardy, a noted English biologist, had just suggested (1912) that since a surface is extremely unsymmetrical with reference to the material on its two sides, the molecules in the surface should be oriented. Thus the theory of dissymmetry and that of solubility gave rise in two different minds to the same suggestion."

Especially in his later papers, Harkins deals extensively with emulsion polymerization, soap micelles, and other matters related to the formation of colloids. He also deals with adsorption and with the surfaces of solids and their interaction with liquids. Again, in the Gibbs Medal address, Harkins explains his interest in atomic nuclei as follows:

"In order to understand the action of surfaces, it appeared essential to learn as much as possible about the electrical structure of molecules and of atoms, so, in 1913, I began to study more intensively the current theories of atomic structure. In 1904, Nagaoka had suggested that an atom consists of a central sun or nucleus and a system of negative electrons as satellites. This theory was amplified by Rutherford, who showed that the positively charged atom nucleus appears to be extremely minute in comparison with the space occupied by the atom. For many years the phenomena of radioactivity had been extremely fascinating to me, and this was undoubtedly what caused my attention to be directed more specially to the nucleus, which de-

termines the stability and even the existence of the atom as a whole."

A series of three papers by Harkins and his student E. D. Wilson in 1915 represents the first of a number of papers published over the years in which Harkins developed ideas on the structure of atomic nuclei. The papers distinguish carefully between chemical elements and atomic species. In general, an element is a mixture of atomic species (isotopes). In 1915 it was already clear that most of the lighter elements have atomic weights very close to a unit that is slightly (about 0.77 percent) less than the mass of the hydrogen atom. The 0.77 percent discrepancy was attributed by Harkins and Wilson (and also independently by Rutherford and others) to what they called a "packing effect," ascribed to a loss of mass predictable from Lorentz' electromagnetic theory if protons and electrons interact at sufficiently close range. They included a speculation that the conversion of hydrogen to helium might be a source of the energy for the sun and stars. Harkins' friend A. C. Lunn, professor of mathematical physics, made the calculations for him. As time went on, it became increasingly clear from mass spectroscopic evidence that those elements whose atomic weights differ from integral multiples of the basic unit are mixtures of isotopes.

Quoting G. N. Lewis [Phys. Rev. 46 (1934):897], "It was Harkins who first called attention to the striking connection between the atomic weights of the elements and their abundance, not only in the earth's crust, but [as a much better sample of the solar system], in the meteors." And it was he who first used these abundances as criteria for the relative stabilities of various atomic species. After E. Rutherford's proof of the nuclear atom, and until the experimental proof by J. Chadwick in 1932 of the independent existence of the neutron, it was generally believed that nuclei are built of protons (not so named at first) and electrons. Harkins noticed that the relative abun-

dances, hence stabilities, of different atomic species are by far greater for nuclei containing an even number of protons and electrons; the next class, in terms of abundance, contains an odd number of protons and an even number of electrons. Two much rarer classes contain an even number of protons but an odd number of electrons, or odd numbers of both protons and electrons.

It was also apparent to Harkins that many of the lighter species (e.g.,  $^{12}\text{C}$ ,  $^{16}\text{O}$ ,  $^{20}\text{Ne}$ ) could be thought of as built of  $\alpha$ -particles; the  $\alpha$ -particle itself, the helium nucleus, being a very stable composite, formed by close packing of four protons and two electrons,  $p_4e_2$ . The atomic weights of the  $\alpha$  composites such as  $^{12}\text{C}$  and  $^{16}\text{O}$  showed little further packing effect. Harkins did not attempt to give a categorical answer to the question of whether such nuclei *consist of* (nearly unchanged)  $\alpha$ -particles, or whether they merely could be *built from*  $\alpha$ -particles. It was of course known that  $\alpha$ -particles can have an independent existence.

In a similar way, Harkins concluded that nuclei such as those of  $^{19}\text{F}$  and  $^{23}\text{Na}$  could be built from, or possibly consist of,  $\alpha$ -particles plus a hypothetical  $\nu$ -particle ( $p_3e_2$ ). Harkins mentioned, but did not emphasize, the possibility of the independent existence of this and other particles (the helion,  $p_4e_4$ ;  $p_2e$ ; the  $\mu$  particle,  $p_2e_2$ ; and a particle  $pe$ ). He mentioned that  $p_3e_2$  and  $p_2e$ , since known, would be isotopes of hydrogen (the triton, and the deuteron). Rutherford entertained similar ideas, and independently (and earlier than Harkins) spoke of packing (of H to form He), but it was Harkins who made the major contributions on the stabilities of atomic species and the structure of nuclei. This work culminated in his "new periodic system" of atomic species expressed in a diagram of "isotopic number" versus atomic number. Here, if the structure of any nucleus is written as  $(p_2e)Z (pe)_n$ ,  $n$  is the isotopic number if  $Z$  is the atomic number;  $n$  was later recognized as a neutron number.

It was first shown by Chadwick in 1932 that the grouping (pe) can exist as a free particle, the neutron. Rutherford in his Bakerian lecture in 1920 describes the properties of the neutron as a conceivable free particle. The idea that such a particle might exist kept Rutherford's laboratory "on the lookout" for it and doubtless thereby contributed to Chadwick's discovery. (A little later, it was realized that, in its formation from a proton and an electron, the neutron undergoes a much more radical change than simple association; for example, while the proton and the electron each have a spin  $\frac{1}{2}$ , so does the neutron. Its spin cannot be accounted for by the otherwise expected vector addition to give a spin 0 or 1.) Rutherford in his Bakerian lecture refers to Harkins' 1920 paper, "The Nuclei of Atoms and the New Periodic System," in which Harkins refers to the (pe) grouping. In 1921, in a paper communicated to *Philosophical Magazine* by Rutherford, Harkins first introduced the word *neutron* to describe this particle.\* Harkins in an article in *Science* in 1946 said "that a neutron exists was assumed independently by Rutherford and Harkins in 1920."

Stimulated by work in Rutherford's laboratory, Harkins undertook work on the disintegration of nitrogen nuclei by the impingement of  $\alpha$ -particles. Here he used C. T. R. Wilson's cloud chamber method to photograph the tracks of the  $\alpha$ -particles and the disintegrating nuclei. Although apparently not the first or the most immediately successful in the use of this method, he and his students made valuable contributions. Later, after the neutron had been discovered, he did extensive work on the disintegration of nitrogen and other nuclei by

\* N. Feather in "A History of Neutrons and Nuclei. I.," *Contemporary Physics* 1(1960):191, "A History of Neutrons and Nuclei. II.," *Contemporary Physics* 1(1960):257, and "Chadwick's Neutron," *Contemporary Physics* 15(1974):565 has reviewed the development of the neutron concept. As he points out, the word neutron was first used by W. Sutherland in 1899 and in 1903 by W. Nernst, but for different concepts than that of the current neutron. Feather refers to Harkins on pp. 260-61 of his earlier papers.

neutrons. In the course of this work, he developed ideas about the details of the mechanism leading to disintegration. He insisted that the neutron (or  $\alpha$ -particle) is first captured and held very briefly in the form of a compound nucleus, which then disintegrates. For some years this formulation was frowned on by the theoretical physicists, but eventually it was seen that he was right.

In his later work, Harkins had some ideas about shell structure in nuclei. Harkins was a pioneer, but he was also quick to take up and implement new ideas originating elsewhere. Soon after Lawrence had invented the cyclotron, Harkins built one at the University of Chicago with the cooperation of his students. Planning began in 1935, and the cyclotron (larger than Lawrence's original) was in operation in 1936. Soon it also began to be used by physicists, Enrico Fermi in particular, for neutron-diffusion studies. With the advent of fission, and Harkins' official retirement in 1939, the cyclotron was turned over to the physics department, under S. K. Allison, a former student of Harkins and afterward a professor of physics. The Harkins cyclotron was used thereafter in the Manhattan Project at Chicago during the war. The papers in Harkins' bibliography make no mention of the cyclotron, except for a brief note in *Science* in 1936 by Moon and Harkins, which shows them as thinking about how to improve on Lawrence's cyclotron.

One activity not yet mentioned is Harkins' work on the separation of isotopes. Except for a partial separation of neon isotopes by F. W. Aston using a diffusion method, Harkins was the first to obtain such a separation. This he and his students accomplished in the case of chlorine, using the diffusion of HCl through clay pipe stems. Afterward there was work (not the first but by far the most extensive) on the partial separation of mercury isotopes.

In conclusion, I quote from T. F. Young's introduction to Harkins' book on surface chemistry, mentioned above. "It is of

interest to note that many of Harkins' contributions [about one-third] were made after his formal academic retirement in 1939. His productivity, especially after 1940, was greatly enhanced by financial aid from industries and government agencies interested in catalysis, lubrication, the production of rubber, and other applications of surface chemistry. It was his own dominating curiosity, however, which was the principal cause of the productiveness which endured literally to the last day of his life."

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- Chem. Rev. = Chemical Reviews  
Colloid Symp. Monogr. = Colloid Symposium Monograph  
Ind. Eng. Chem. = Industrial and Engineering Chemistry  
J. Am. Chem. Soc. = Journal of the American Chemical Society  
J. Biol. Chem. = Journal of Biological Chemistry  
J. Chem. Phys. = Journal of Chemical Physics  
J. Colloid Sci. = Journal of Colloid Science  
J. Franklin Inst. = Journal of the Franklin Institute  
J. Phys. Chem. = Journal of Physical Chemistry  
J. Phys. Colloid Chem. = Journal of Physical and Colloid Chemistry  
J. Polym. Sci. = Journal of Polymer Science  
Philos. Mag. = Philosophical Magazine  
Phys. Rev. = Physical Review  
Proc. Natl. Acad. Sci. = Proceedings of the National Academy of Sciences  
Publ. Am. Assoc. Adv. Sci. = Publications of the American Association for  
the Advancement of Science  
Sci. Mon. = Scientific Monthly  
Z. Phys. = Zeitschrift für Physik

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