BIOORGANIC CHEMISTRY 20, 269–284 (1992)

TRIBUTE

Frank Henry Westheimer: The Celebration of a Lifetime in Chemistry

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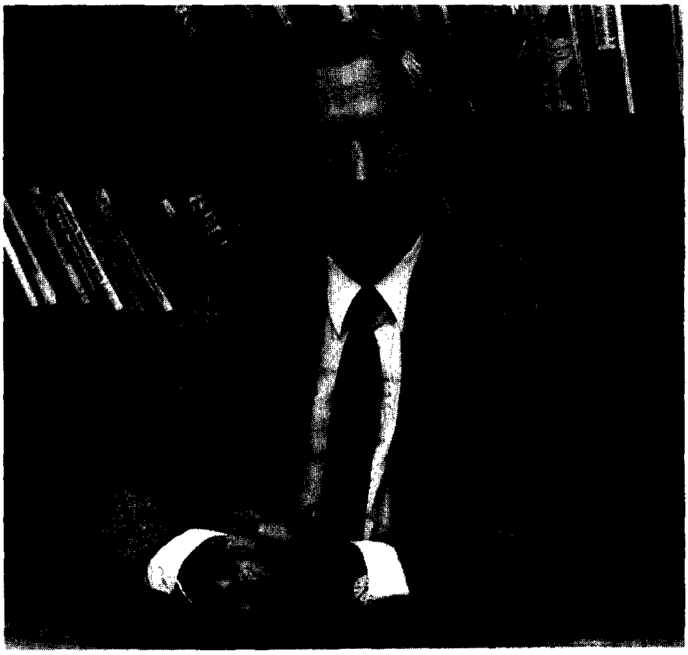
Received June 6, 1992



It is not usual for the literature to record an appreciation of the work of one of its greatest living scientists: sadly, that privilege is too often the preserve of the dead. But Frank Westheimer is not a usual kind of man, and this short article attempts to place on the public record something of his great love for and contribution to chemistry over a period of nearly 60 years. By way of justification, if indeed any justification were needed, I believe the young and not-so-young may benefit from an exposure to this story now, and not at some distant future time.

On 15 January 1992, he turned 80, and the science of chemistry and the closely related disciplines of physical organic chemistry, biorganic chemistry, and biochemistry can be grateful first for the quality of his genes. In 1937, he married

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Jeanne, who has supported him and their family throughout with an easy and consummate grace born of great happiness. Science can also be grateful for the quality of her genes.

To mark his 80th birthday, a "group meeting" was organized in San Francisco so that it overlapped the meeting of the American Chemical Society. A splendid banquet at the St. Francis Hotel on the evening of Saturday, 4 April, saw the gathering of 60-odd former students, postdoctorals, wives, husbands, and a handful of ring-ins who met to honor their good friend and mentor. If there was a coherent message in the after-dinner meanderings, it surely was that one does not need to be 80 to have entered upon the years of anecdotage! In his simple response, Westheimer's mind turned to chemistry: this time he was concerned about the solution of another significant problem of modern chemistry—manic depression.

A special symposium of the American Chemical Society was held on the following Monday morning in honor of his receipt of the Repligen Award for the Chemistry of Biological Processes, probably the first award specifically to recognize his enormous contribution to biochemistry. Four papers were presented by former students, and at the end, Professor Westheimer allowed himself the luxury of some reminiscences to which I shall briefly return.

Born in Baltimore, Westheimer graduated B.A. *summa cum laude* from Dartmouth in 1932, M.A. from Harvard in 1933, and Ph.D. from Harvard in 1935 for research done under James Bryant Conant for some 6 months before he formally assumed the Presidency of the University on 5 September 1933, and then under Elmer P. Kohler. After a postdoctoral stint with Louis Hammett at Columbia (1935–1936) and an independent Research Associateship at the University of Chicago (1936–1937), he joined the faculty at the University of Chicago as an Instructor in 1937 and rose through the ranks to Professor in 1948. He held this position until he accepted a Visiting Professorship at Harvard (1953–1954), "and stayed on." In 1960, he was appointed to the Morris Loeb Chair at Harvard, and in 1982, at the age of 70, became a Senior Professor. He formally retired in 1983, when his teaching commitments, but not his research, also came to an end. He gave up his laboratory in 1988, and is currently Morris Loeb Professor Emeritus.

During the early years of World War II, he undertook classified research, and later served as Research Supervisor in the Explosives Research Laboratory of the National Defense Research Committee (NDRC, Bruceton, PA, 1944–1945). It was during this time that he hung by his finger tips for a very long 5 minutes from an external high second floor window ledge until rescued from a fire in the laboratory.

Over the last 60 years, his contribution to chemistry has been characterized not only by the breadth of its sweep, but also by the sheer brilliance of his vision. For Westheimer is no narrowly defined organic chemist, or physical chemist, or whatever, but a chemist's chemist who is an ornament to our profession: he has made major contributions in most areas. A chronicle of his more than 200 papers is a chronicle of achievement far greater than that which is the lot of most. Indeed, in the fields of physical organic chemistry and bioorganic chemistry, it is a chronicle of unparalleled excellence.

From his earliest work, he has been concerned with the corpus of theory and experiment which constitutes physical organic chemistry. In his first published paper (*1*),[1] he was already considering the "relative reactivities" of various carbonyl compounds, and while a National Research Fellow in Louis Hammett's laboratory, determined the mechanism of the benzilic acid rearrangement (*3*, *4*). At the hoary age of 25, he reported his first "model research in the enzyme field"—the amino acid catalysis of the mutarotation of glucose (*5*). The words "model research in the enzyme field" are definitional of Westheimer: his writing has at once a simplicity and felicity of phrase, and is marked by an admirable economy of words.

In these formative years, such was the paucity of secure knowledge of catalysis that he spent a deal of time developing the tools appropriate to a mechanistic attack on enzymes. Four classical papers (*7–10*; see also *25*) appeared in 1938–1939. In these, Kirkwood and Westheimer, together with Shookhoff, developed the theory of the electrostatic influence of substituents on the dissociation constants of organic acids, a subject of fundamental significance in enzymology as he later established with unassailable clarity (*138*, *143*, *149*, *150*).

Definitive quantitative experiments on the electrostatic influence of substituents, of resonance, and of solvent on reaction rates followed (*11*, *12*, *15–17*, *19*). Also in 1938, he established *specific* amine catalysis (*6*, *13*), a result of fundamental significance in a number of areas, but especially in the mechanism of action of the enzyme, acetoacetate decarboxylase (*72*, *75*, *88*, *93*, *108–111*, *116*, *126–130*, *134*, *138*, *141–143*, *149*, *150*, *153*, *159*).

In 1943, he published the first result of his extended foray into the mechanism of chromic acid oxidations (*20*, *27–29*, *32*, *38*, *42*, *43*, *47*, *56*, *64*, *70*, *78*, *79*, *87*, *90*, *95*). The masterly review (*32*) published in 1949 is eloquent testimony to his ability rapidly to synthesize apparently discordant data and logically to draw far reaching conclusions: his reviews are never a dull recitation of references. This field was further explored by Jan Rocek, who enjoyed the hospitality of Westheimer's laboratory when he first immigrated to the United States from Europe (*87*, *90*).

Work for the NDRC on the nitration of aromatic compounds in sulfuric acid (*21*) and on the mechanism of "oxynitration" of benzene (*23*, *26*) appeared at the end of World War II. The investigation of "oxynitration" led to further definitive studies on the mechanism of mercuration of aromatic compounds (*37*, *41*, *98*).

The year 1946 saw the first paper on the theory of racemization of optically active derivatives of diphenyl (*22*). In this paper with Joseph Mayer, Westheimer laid the ground rules for his entry into what would now be called the field of molecular mechanics. Again the invention was developed in a classical series of articles (*24*, *31*, *34*, *35*, *55*).

Already we see here a distinguishing feature of this man's science—he likes "going on to new things." It is certainly true that he has never fully exploited any of his well-sown fertile fields, but those many who have come after have reaped the better harvest for his labors. It is idle for us to speculate whether this has thereby diluted his overall contribution in any way: as he says in one of his

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[1] Reference numbers refer to the detailed bibliography which appears at the end of the article.

(unrelated) literature rejoinders (*25*), "a value judgment [such as this] is a personal matter, and therefore scarcely a subject for scientific discussion."

In 1949, with Daniel Koshland, Jr., Westheimer established the (accepted) mechanism of the alcoholic fermentation of glucose (*30, 36*), and the same year saw the communication of his initial essay into the chemistry of metal-ion catalyzed decarboxylations (*33*). The full paper (with Rudolph Steinberger) (*39*) signals his subsequent studies in this area (*49, 51, 54, 58, 74, 131*). The paper which involves the determination of "pH-rate profiles" and the spectrophotometric investigation of intermediates is an experimental landmark in physical organic chemistry. It must have been no small source of satisfaction to him to hear in the Repligen Symposium that the detailed results of Koshland and his co-workers[2] in a different system are completely in line with his thinking of 40 years ago. Further, his proposition (*49*) that "metal-ion catalysis can perhaps be described as superacid catalysis in neutral solution" has itself been the catalyst for hundreds upon hundreds of developments in physical organic chemistry, bioinorganic chemistry, and metalloenzymology. His basic proposition which to this day is frequently not properly understood (because of the enormously different electric fields of a proton and a metal ion) is nevertheless simple: metal-ion catalysis can operate under conditions where catalysis by protons is not available.

But perhaps the work for which he is best known in enzymology concerns the direct transfer of hydride ion to the coenzyme NAD^+ from various substrates, a reaction which occurs in some enzyme-catalyzed reactions. In these experiments, he collaborated with Birgit Vennesland, a biochemist who was then also at the University of Chicago (*40, 44–46, 48*). Such was the quality of this exacting exercise that the added bonus of the stereospecificity of the transfer fell into their laps. Once again, the enzymatic experiments were supported by model studies (*50, 61, 63, 66, 83, 86, 102*).

In 1955, he published the first article in his monumental study of the chemistry of phosphate esters and other phosphorus derivatives (*51, 52, 57, 65, 67–69, 73, 84, 96, 99, 104–106, 112–115, 117–120, 122–125, 132, 136, 137, 139, 140, 148, 152, 154–157, 160–166, 168, 171, 174–178, 180, 183, 184, 188–190, 192–197, 202, 206, 209*). In these papers, he introduced "the hypothetical ion," monomeric metaphosphate (PO_3^-), *quantitatively* to account for the mechanism of hydrolysis of monoesters of phosphoric acid. Chemistry was to wait some time to see the proof, and again at his hands, first with the preparation of the very highly reactive monomeric methyl metaphosphate, clearly identified by the products of electrophilic substitution on *N*,*N*-diethylaniline (*157, 160*), and finally of monomeric metaphosphate itself (*117, 190*). That highly reactive intermediates are involved in these latter studies would appear to be beyond doubt: it is the question of the solvation of these species which has attracted its share of comment. Thus Bill Jencks[3] has argued that monomeric metaphosphate is virtually never "free" in solution, but as Westheimer himself has pointed out (*192*), neither is the proton, and Jencks's

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[2] KOSHLAND, D. E. JR., STODDARD, B., LEE, M. AND DEAN, A. (1992) *Abstracts, 203rd A.C.S. National Meeting*, Part 1, *Biol. Chem.* Paper 1.

[3] JENCKS, W. P. (1980) *Acc. Chem. Res.* **13**, 161, and references therein.

contention may obscure the larger truth. Similarly, whereas racemization produced in an appropriate enzymatic reaction would provide relatively unequivocal evidence for "free" metaphosphate, inversion in no way guarantees a simple displacement mechanism.

No less significant was his use of pseudorotation[4] to account for the hydrolytic chemistry of phosphate esters (*84, 117, 118*). The work led to a number of risky predictions[5] which have since been borne out by experiment (*122–125, 132, 136, 137, 139, 140*). This group of papers alone must rank as among the most elegant in the whole of physical organic chemistry.

Studies on the mechanism of thiamin-catalyzed reactions—both enzymatic and nonenzymatic (*53, 62, 71*)—were followed by definitive studies on the mechanism of action of acetoacetate decarboxylase (*72, 75, 88, 93, 108–111, 116, 126–130, 134, 138, 141–143, 149, 150, 153, 159*). In spite of the fact that no assistance was obtained from X-ray crystallography in this work, the mechanism of action of few enzymes is better understood.

In 1962, Westheimer reported on what is now known as photoaffinity labeling (*85*), with a communication on the photolysis of diazoacetylchymotrypsin. A fuller account appeared in 1966 (*107*), but the first clear signs of the success of the invention came when independent work began to appear in 1969. Again, he went on to develop the ideas (*133, 135, 145–147, 151, 166, 167, 169, 170, 172, 181, 185*). Undoubtedly the 1962 report was a long way ahead of its time. Many hundreds of articles have since appeared, and the noteworthy range of these studies is taken up by the inventor in his last papers on these systems (*172, 181, 185*). Science is surely now in a position to rate this as one of the great inventions of physical organic chemistry.

Finally, it would be completely delinquent of me not to mention the many papers which do not carry his name that report work done by students, postdoctorals, and visitors in Westheimer's laboratory. Bender's tetrahedral intermediate readily springs to mind.[6] In many of these papers, his was the strong guiding hand, so that if he was not the *de jure* father of the work, he was certainly more than the caring midwife who attended its birth.

Enough chemistry for the present. Frank Westheimer is also an exemplary teacher, and whether in a formal lecture (as in his introductory organic chemistry lectures at Harvard), in a seminar, or in a one-to-one situation, his clarity of presentation, formidable logic, and encyclopedic knowledge ensure his audience a treat; this man will discuss chemistry with anyone at any level, if they are prepared to listen. The Friday lunchtime group gatherings in my day have a special place, and here it would be impolitic to try to remember how many times Frank's lunch wrapper managed to hit the trash bin with or without a cannon off the cush. Most remember the Bartlett–Westheimer group seminars with some trepidation, but with great affection. During his life, he has held many visiting Fellowships and

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[4] Berry, R. S. (1960) *J. Chem. Phys.* **32,** 933.

[5] Popper, K. R. (1969) Conjectures and Refutations, 3rd ed., p. 36 et seq, Routledge and Kegan Paul, London.

[6] Bender, M. L. (1951) *J. Am. Chem. Soc.* **72,** 1626.

to date has delivered some 20 distinguished lecture series worldwide, from the Harrison Howe Lecture of the University of Rochester (1954), to the Morrell Lectures of the University of Cambridge (1962), to the Priestley Lectures of Pennsylvania State University (1968), to the Ingold Lecture of the Royal Chemical Society (1983), to the Hitchcock Lectures of the University of California at Berkeley in 1987. He has so far made only one visit to Australia, and that was in 1969 when he was invited to give the CSIRO's prestigious Fourth David Rivett Memorial Lecture in Canberra (*138*), the first American so called, and the first ever to speak on chemical research.

However, his path has not always been easy. For the special benefit of the young and to ensure a balanced coverage, I report that he was once nominated for a Visiting Professorship at the University of Queensland, a position which required the recipient to give some six or more lectures and to discuss research with the graduate students: such is the quality of our selection procedures that he failed to make the grade! One recalls President Lowell's words—"The fastest way to ruin a University is to fill it with good men."

Professor Westheimer has also been active in university, scientific, and public affairs for many years. He was Chairman of the Harvard Chemistry Department from 1959 to 1962, and after a sabbatical in the other Cambridge, went on to chair the Committee of the National Academy of Sciences on the Survey of Chemistry (1964–1965). Their report, known universally as the Westheimer Report (*100, 101*), provided the first real assessment of chemistry in the affairs of the people of the United States: it has undoubtedly served them well. Indeed, it is still an appropriate model for those in other climes who would seek to make similar inquiry.[7] He was a Member of President Johnson's Scientific Advisory Committee (1967–1970), and has had two terms on the Council of the National Academy of Sciences (1973–1975, 1976–1978). He served as a Councillor of the American Philosophical Society from 1981 to 1984, and as Secretary of the American Academy of Arts and Sciences from 1985 to 1990.

His devotion to teaching goes much deeper than the lecture hall. He is now increasingly concerned about the education of American youth. Since President Lowell in 1910 replaced Eliot's free elective system with one of "concentration and distribution," Harvard has been at the center of the debate on what constitutes an appropriate liberal education. Conant's presidency was plagued by this very problem for many years.[8] When the Harvard faculty in May 1978, by a vote of greater than three to one, opted for a "core curriculum," Westheimer came out fighting (*204, 205*). He argues that because the "core" minimizes science (2 half-courses of 16 full courses, 6%), most of the Harvard nonscience graduates are uneducated since they have virtually no science (*210*). He brings a new dimension to the debate and his propositions have a significant urgency. All universities worthy of the name would do well to consider them carefully. With the same unassailable logic that he used in the application of pseudorotation to phosphorus

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[7] For his own views on the Report some 17 years later, see *Chem. Eng. News* (1983) **61,** 14–20.

[8] Conant, J. B. (1970) My Several Lives, Harper & Row, Inc., New York. See, for example, Ch. 3, pp. 20–26; Ch. 27, pp. 363–373.

chemistry, he points out that learning in science is *necessarily* vertical (sequential), whereas learning in the humanities is, with the notable exception of language instruction, much more horizontal. Thus one cannot take up a course on quantum mechanics without the necessary prerequisites. In his Priestley Medal address (*207*), he develops the ideas further, and it should be compulsory reading for all university administrators.

Such distinction and downright hard work should be rewarded, and with perhaps one too obvious exception, there is scarcely a major honor that he has not received. He holds honorary Doctorates of Science from Dartmouth (1961), Chicago (1973), Cincinnati (1976), Tufts (1978), North Carolina and Bard College (1983), the Weizmann Institute of Science (1987), and the University of Illinois at Chicago (1988). He was elected to Membership of the National Academy of Sciences and the American Academy of Arts and Sciences in 1954, to Fellowship of the American Association for the Advancement of Science in 1976, to Foreign Membership of the Royal Society of London in 1983, and to Honorary Membership of the New York Academy of Sciences in 1985.

For his work during the Second World War, he received the Army–Navy Certificate of Appreciation and the Naval Ordnance Award in 1946. His major contribution to chemistry was just beginning to be recognized when he was awarded the Willard Gibbs Medal in 1970. The James Flack Norris Award of the American Chemical Society came in the same year, followed by the Theodore William Richards Medal in 1976. Honors came in abundance in 1980–1983: the National Academy of Sciences Award in Chemical Sciences (1980), the Richard Kokes Award of The Johns Hopkins University (1980), the Charles Frederick Chandler Medal of Columbia University (1980), the Lewis S. Rosensteil Award of Brandeis University (1981), the Robert A Welch Award from the Robert A Welch Foundation (1982), the Arthur C. Cope Award of the American Chemical Society (1982), the William H. Nichols Medal of the New York Section of the American Chemical Society (1982), and the Sir Christopher Ingold Medal of the Royal Society of Chemistry, London (1983). Then followed the Wheland Medal of the University of Chicago (1986), the University Medal of the University of Illinois at Chicago (1986), the National Medal of Science (1986), and the Paracelsus Medal of the Swiss Chemical Society in 1988. It was also in 1988 that he was elated to receive the highest award the American Chemical Society can bestow—the Priestley Medal for distinguished service to chemistry (*207*).[9]

In his closing remarks at the Repligen Symposium, he acknowledged the Repligen Award with pride, but went on to reveal a little of the warmth and essential modesty that endear him to everybody by suggesting that the greatest of all his awards is the group of young men and women who came to work with him and to share his love of chemistry. Characteristically, he also remembered those who have already passed on.

Perhaps it would not be improper to end on a personal note. In the Solera libretto of Giuseppe Verdi's *Nabucco*, there is a magnificent line where the chained Hebrew slaves sing in chorus, "Va, pensiero, sull'ali dorate," and it has never

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[9] See also RAWLS, R. L. *Chem. Eng. News*, **65**, 21–23 (1987).

been far from me during the writing of this piece. I can only hope that the young in chemistry are inspired by his example as we were and are, and that something of the joy known by the highly privileged band of Westheimer's people comes through.

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