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The Scientific Work of Saul Winstein

Saul Winstein was born in Montreal, Canada, October 8, 1912. He came to the United States in 1923, graduated from high school in Los Angeles, and attended the University of California at Los Angeles, obtaining the A.B. in 1934 and the M.A. in 1935. He received the Ph.D. in 1938 at the California Institute of Technology. After two postdoctoral years and a year as instructor at the Illinois Institute of Technology, he returned to UCLA where he was Professor of Chemistry from 1947 until his sudden death in 1969, at the age of 57 and at the height of his career.

The years covered by Saul Winstein's publications (from 1933 to 1971) brought a greatly expanded use of physical methods as tools in organic chemical research and of physical reasoning in the interpretation of organic reactions.

He was trained in careful and inventive experimentation on synthetic materials chosen to illuminate the nature of chemical reactions. Eight papers arising from Winstein's undergraduate and master's degree work with William G. Young concern the quantitative study of the isomerism and isomerization of allylically related bromides and Grignard reagents. These systems continued to intrigue Winstein and he returned to them in 1942 (18), 1949 (54), 1951 (55), 1960 (144 and 159), and 1963 (186).

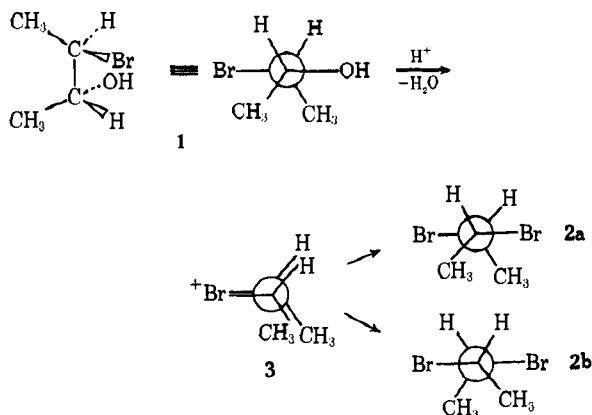
The period of Winstein's graduate work was one of important worldwide activity in reaction mechanisms. Hughes and Ingold in London had classified solvolyses into S_N1 and S_N2 types by their overall kinetic order with respect to nonsolvent nucleophiles. Hammett and coworkers at Columbia had demonstrated the polymolecular role of solvent in solvolytic reactions. A. R. Olson and coworkers at Berkeley had supported the absence of any free ions even in S_N1 solvolysis. Stereochemical inversion in displacement reactions had been generalized by Polanyi and coworkers at Manchester. In his first major independent paper (13), written during a postdoctoral year at California Institute of Technology, and with an acknowledgment of the help and advice of Linus Pauling, Winstein discussed the solvolysis of *tert*-butyl chloride in terms of polymolecular solvent participation and a largely covalent attachment of an alcohol or water molecule to the carbonium ion center. This paper shows that he was greatly influenced by the current literature and was

extending his concern with the fusion of kinetics, stereochemistry, and mechanism. He must have been deeply dissatisfied with the simplism of this and other thinking of the times about solvolysis, for much of his later effort went into the study of just how complex a reaction solvolysis is.

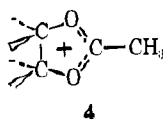
Of the six papers growing out of his doctoral work with Howard J. Lucas, five foreshadowed Winstein's later work in an important way. Three papers (10, 11, and 15) on the stereochemistry of displacements involving 3-bromo-2-butanol opened the entire field of neighboring group participation. At the same time studies of the coordination of silver and mercuric ions (9 and 16) with unsaturated compounds provided prototypes of three-center, two-electron bonds and initiated an interest in coordinative as well as covalent organometallic reactions which showed itself in twelve later papers extending to 1969. The largest group of these dealt with organomercurials (89, 90, 99, 179, 180, and 186); there were also contributions to metallic oxidations and reductions (56-58, 60-67, 135, and 182), one paper on carbenoids (279), and in recent years use of metal carbonyl complexes to induce and detect homoaromatic character in cyclic polyenes and their ions (224, 233, 247, and 284).

In Winstein's thesis work it was shown that in the replacement of the OH by Br in 3-bromo-2-butanol, the requirements for a double inversion involving the bromine atom neighboring the OH were uniquely met. *erythro*-3-Bromo-2-butanol led to *meso*-2,3-dibromobutane with overall retention of configuration. Simple front-side attack of bromide ion on the protonated alcohol would have required that optically active *threo*-3-bromo-2-butanol (**1**) should yield optically active 2,3-dibromobutane (**2a**). The fact that **1** actually yielded diastereomerically pure but totally racemic (**2ab**) was uniquely consistent with the participation of the *meso* intermediate (**3**) formed in an intramolecular displacement with configurative inversion and reacting with a second inversion at either carbon atom. During the next few years widening implications of this discovery were worked out.

On the second try (!) Winstein was awarded a National Research Fellowship in 1939-1940; at Harvard he explored the reaction of vicinal dibromides with silver acetate, which led to the discovery that a neigh-



boring acetoxy group is an excellent intramolecular nucleophile, participating in displacement reactions with the formation of a 5-membered cyclic dioxolenium or acetoxonium ion (4). Unlike the bromonium ion,



this intermediate could be detected not only by the overall stereochemical course of the reaction but by trapping with water, which attacked the carboxy carbon, leading to ring opening without inversion, or by alcohol, which led to an orthoester that could be independently synthesized (Winstein and Buckles, 19, 20, 24; Winstein, Hess, and Buckles, 23). Years later Winstein and Friedrich (190) isolated an acetoxonium fluoroborate.

Back at UCLA on the faculty after a year as instructor at the Illinois Institute of Technology, Winstein began attracting students and accelerating his research. In varying degrees, involvement in displacement reactions could be demonstrated for hydroxyl and its negative ion, methoxy and other ether groups, amido groups as analogs of the acetoxy, and the carbon-carbon double bond, which is nucleophilic by virtue of its π electrons. The carboxylate ion neighboring group, which had been regarded by Ingold and co-workers¹ as having an *electrostatic* configuration-stabilizing effect on the cation, was now regarded as forming a covalent α -lactone and reinvestigated in the context of other neighboring group participations (Winstein and Grunwald, 48).

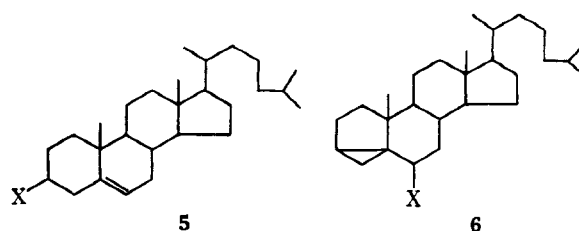
In 1944 and 1945 Winstein's publication program suffered an interruption in common with those of most American academic chemists who had an opportunity to contribute to the pressing national problems of the time. A series of papers (28-42) in 1946-1948, in collaboration with T. L. Jacobs and a number of co-workers, describes the synthesis of a series of naphthalene and quinoline amino alcohols for testing as antimalarial drugs under a program of the Office of Scientific Research and Development. This work shows little connection with earlier or later Winstein chemistry, except that halohydrins and epoxides were developed as superior intermediates in the preparation

(1) W. A. Cowdrey, E. D. Hughes, C. K. Ingold, S. Masterman, and A. D. Scott, *J. Chem. Soc.*, 1252 (1937).

of β -aminoethanols, replacing the former preparation and reduction of β -amino ketones.

The flow of fundamental research was resumed in 1946 with an important half-page communication by Winstein and Grunwald (27) reporting that neighboring group participation, demonstrated by the stereochemical criteria, is often accompanied by great acceleration of the ionization process. The documentation and discussion of this effect, with Grunwald, Buckles, Hanson, and Ingraham (44-46), gave a precise definition of the *driving force* due to participation and provided ways to determine its value by comparing the observed rate with that to be expected on the basis only of the effect of the substituent on unassisted ionization rate. In these papers the nature of solvolysis of arene-sulfonates was also carefully analyzed, and the advantages of sulfonate acetolysis in the study of carbonium ion reactions were established.

The participation of a neighboring double bond in a displacement reaction (Winstein, Adams, Walborsky, Schreiber, and Shatavsky, 47, 60, 91, and 93) raised questions that could not be answered by analogy with the bromonium ion. In behaving as a participating neighboring group, did the π electron pair of the double bond remain associated with both carbon atoms, or did the three-center structure that this would imply give way to a carbonium ion with localized charge on one carbon and a covalent bond to the other? The work of Winstein and Adams (47) showed that the participation of the double bond of a cholesterol derivative (5) to yield a derivative (6) of *i*-cholesterol has a dra-



matic counterpart in the rapid reverse rearrangement (Winstein and Schlesinger, 57) in which the "saturated" cyclopropyl ring of 6 acts as a participatory group more powerful than the double bond. Since the speed of the rearrangement in both directions could not mean that the reaction was exothermic in both directions, a mechanism had to be sought whereby the barrier to ionization was lowered by charge delocalization involving either kind of neighboring group. This would occur if a common intermediate ion with special charge delocalization were directly formed from either structure, 5 or 6. The analogy to the familiar allylic systems was only reinforced by further experimental study, and soon the concept of "homoallylic" interactions was used [with Simonetta (79), Kosower (107, 108), Allred and Sonnenberg (140, 146), Piccolini (174), Birladeanu, Hanafusa, and Johnson (229), Gasic, Whalen, Johnson, and Jones (255, 256), and Poulter (270)] as a unifying framework for the enhancement of solvolytic reactivity by a 3,4 double bond and by a 1-cyclopropyl substituent. Theoretical considerations (79) indicated that these interactions in both directions should be enhanced by structural factors favoring close approach of the overlapping orbitals (the p orbital at

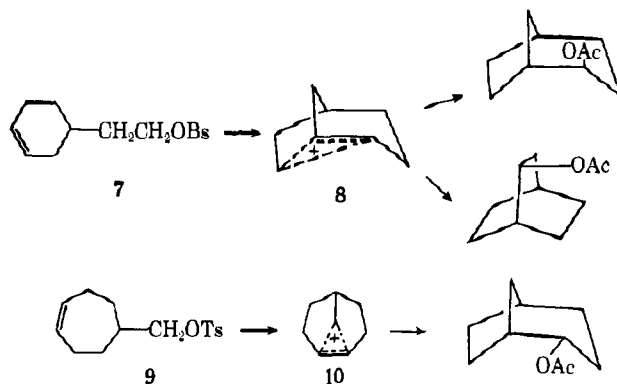
C₁ overlapping the 3,4 double bond in σ fashion, or the α -cyclopropyl group in π fashion).

Participation of neighboring phenyl, reported by D. J. Cram in 1949, provided a flexible series of "homobenzyl" structures. Depending upon the solvent, substituents in the aromatic ring, and variation from primary to tertiary at C-1 and C-2, these compounds were found to react without participation or with total participation of the neighboring aryl group (with Schreiber, Corse, Marshall, Morse, Grunwald, Jones, Trifan, Brown, and Schlesinger, 68-73; Baker and Smith, 193, 194; Leute, 240; Diaz and Lazdins, 263, 272).

In intermediate cases the solvolyses of primary (263) and secondary (272) β -aryl-alkyl toluenesulfonates were shown to be resolvable into an aryl-assisted mechanism and an independent mechanism without intramolecular assistance. Most of the study of such reactions was by systematic observations of the effect of structure on rate and product, but in a few cases it was possible to trap a phenonium ion in the form of a spirooctadienone (e.g., Winstein and Baird, 125, 181) and in others to observe the phenonium ion directly by nmr in low-temperature superacid solvents such as those used extensively by G. A. Olah (with Ebersson, 217; Brookhart, Anet, and Cram, 238).

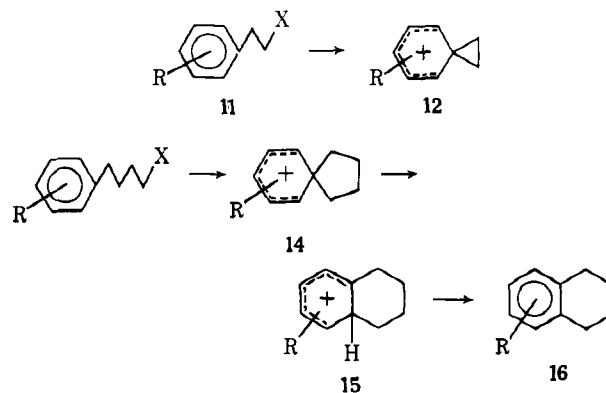
The expanding list of nucleophilic neighboring groups capable of participating in displacement reactions gave little warning that under special circumstances saturated groups might show similar behavior. However, as early as 1939 C. L. Wilson and coworkers had suggested a bridged ion as the reason for the stereospecificity of the camphene hydrochloride-isobornyl chloride rearrangement. Modifying the isborneol structure so as to replace structural isomers by enantiomers, Winstein and Trifan (58, 73, 74) observed that C-6 of *exo*-norbornyl *p*-bromobenzenesulfonate could imitate the behavior of neighboring bromine both in enhancing ionization rates and in controlling the stereochemistry of the solvolytic process. For this kinetic and stereochemical influence of neighboring groups the general term "anchimeric" was introduced in 1953 (77). Systematic exploration showed that both saturated hydrocarbon groups and hydrogen atoms are weak participants of this sort [e.g., with Gadiant, Stafford, and Klinedinst (134), with Clippinger, Howe, Vogelfanger, Colter, Friedrich, and Holness (206-208)], and become strong ones only in molecules having strain that is relieved at the transition state. Winstein never underestimated the role of steric factors in these phenomena, but neither could he be content with fragmentary steric explanations. He took some pains to synthesize new structures in which the mutual influence of steric strains and neighboring group participation could be traced and evaluated (e.g., 134; Winstein and de Vries, 149; Winstein, Thompson, and Bruck, 150, 153; Winstein and Hansen, 160, 161; Winstein, Carter, and Howe, 211).

The observation of Winstein and Carter (171) that the intermediate ion (8) characteristic of solvolysing *exo*-bicyclo[3.2.1]octyl-2-tosylate could be generated from Δ^3 -cyclohexenylethyl bromobenzenesulfonate (7), distinct from the corresponding *endo* ion 10, generated by LeNy² from Δ^4 -cycloheptenylmethyl tosylate (9), established a connection between π participation by a



carbon-carbon double bond and σ participation by a neighboring saturated group. Thus the "nonclassical" ion structure through which a neighboring saturated group participated in an ionization was the same as, or closely related to, that through which a more remote double bond gave participation. The participation of neighboring groups with, and those without, basic electron pairs were linked together as aspects of the principle of optimal charge delocalization through the most favorable overlap of atomic orbitals.

Whereas with $7 \rightarrow 8$ and $9 \rightarrow 10$ both carbon atoms of the double bond appeared to be bonded in the ion, a participating aryl group achieved its best delocalization of charge in the pentadienyl unit of the phenonium ion. While the strain in 12 enables it to open solvolytically to give only displacement products of 11, the ion 14



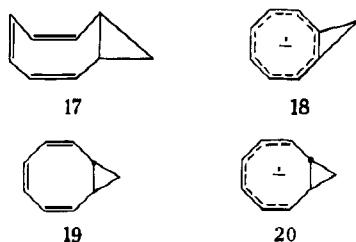
has so little strain that it is not attacked by solvent. Both 2,4-dimethoxy substitution, activating Ar₁ of 13, and 3,5-dimethoxy substitution, activating Ar₂, lead to the tetralin 16 in the anchimerically assisted portion of the solvolysis (Winstein and Heck, 120, 121; see also 97, 123).

Special structures that place a double bond (91, 93, 153, 161; Winstein, Diaz, and Brookhart, 231, 232) or a cyclopropane ring (with Sonnenberg and deVries, 143; with Battiste, Haywood-Farmer, Malkus, and Seidl, 282) in spatial proximity to an ionizing center were observed to exhibit large driving forces (up to factors of 10¹¹ in rate) toward ionization.

The similarities and differences between a double bond and a three-membered ring with respect to conjugation and charge delocalization fascinated Winstein and stimulated his inventiveness. He conceived an entire "homoaromatic" series of compounds having part or all of the classical double bonds replaced by cyclopropane rings. Because such intervention of a

(2) G. LeNy, *Compt. Rend.*, 251, 1526 (1960).

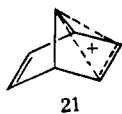
three-membered ring introduces a link of σ orbital overlap in the plane of the conjugated system, the occurrence of homoaromatic properties is always subject to a balance of forces that could not be predicted. In a series of explorations of potentially homoaromatic systems (142, 143, 163, 164, 183, 184, 188, 189, 205, 236, 239, 248–251, 260, 269) it was found that none of the neutral molecules exhibited enhanced stabilities or ring currents, but that a number of isolable cations, anions, and anion radicals showed striking evidence of a homoaromatic ring current. Low ionization potentials of the neutral species also indicated that the cation radicals were appreciably stabilized. In the course of this work some tractable and interesting metal ion complexes of homoaromatic ions were prepared and studied (219, 224, 226, 233–235, 284). By the choice of metal, the same ion could be made into a homoaromatic or a hexatrienoid complex: the free cyclic ion $C_8H_9^+$ and its six- π -electron metal carbonyl complexes, $C_8H_9^+Mo(CO)_3$ and $C_8H_9^+W(CO)_3$ were shown to be aromatic homotropylium species, while the corresponding four- π -electron iron tricarbonyl complex was a "classical," nonaromatic one (with Kaesz, Kreiter, and Friedrich, 219). An elegant example of stereoelectronic control of homoaromaticity was found in the contrast between the homoaromatic anion radical **18** generated at low temperatures from cis-fused bicyclo[6.1.0]nonatriene **17** and the entirely



different anion radical **20** of the hexatriene type from the trans-fused isomer **19** (with Rieke, Ogliaruso, and McClung, 234; with Moshuk and Petrowski, 260). This was recognized as an example of the operation of the Woodward–Hoffmann rules: the aromatization of the cis-fused triene is analogous to the allowed disrotatory ring opening, while the corresponding process in the trans-fused triene would be forbidden.

The work on homoaromaticity was reviewed in 1967 and 1969 (239, 269).

Dynamic nmr studies with Lustgarten and Brookhart (252, 253), combined with deuterium and methyl labeling, made possible a detailed account of the rearrangements and barriers in the delocalized, unsymmetrical 7-norbornadienyl cation.³ In fluorosulfonic acid the unsymmetrical structure **21**, in which one pair



of ethylenic hydrogen atoms is clearly differentiated from the other, is lower in energy than the classical ion or the symmetrical transition state between the equivalent unsymmetrical structures by at least 19.6 kcal/

mol. More rapid than flipping of the bridge is a rearrangement into and out of the bicyclo[3.2.0]heptadienyl ring system, resulting in a complete scrambling of the unbound ethylene pair with the bridge and bridgehead protons. Even substitution of a methyl group at C-7 only lowers the bridge flipping barrier by about 7.2 kcal/mol, while it favors rearrangement into a new charge-delocalized cation with methyl at C-2 and the methylated double bond being the one engaged with the bridge carbon.

Because solvolysis was the most flexible system for studying the chemical behavior of carbonium ions, it received much attention in the early years. While much important behavior centered around the structure of carbonium ions, more and more phenomena were discovered that depended critically upon cation–anion pairs of various degrees of tightness. The discovery of these effects was accelerated by the fact that acetic acid was a very useful solvent for solvolytic studies; because of its low dielectric constant, strong solvation of anions, and weak basic and nucleophilic character, it revealed some ion-pair effects not seen nor suspected in aqueous or alcoholic solvents.

Internal return was observed in solvolytic reactions where an identical ion pair is produced from two structural (55, 75, 76) or stereo (74) isomers. The evidence consisted of the demonstration that the various nucleophiles present have different relative reactivities toward the carbonium ion at different stages of solvolysis. The recovery of rearranged or racemized starting material, e.g., chloride or sulfonate, in a fixed proportion to the solvolysis product, showed that the original anion had an "inside track." There must be an intermediate in the solvolysis best described as a "tight ion pair," where covalent bonding has given way to ionic, but with so much attractive force between the partners that they may still recombine faster than they are displaced or separated by further solvation and diffusion.

A second effect, never predicted by theory, is the *linear normal salt effect*. Lithium perchlorate in many solvents of low dielectric constant produces linear acceleration of the ionization process up to concentrations far beyond the limiting region where the dilute approximation of the Debye–Hückel theory applies. The magnitude of this acceleration is a rather specific property of the solvent and the salt (Winstein, Smith, Darwish, and Friedrich, 139, 191); ether, a very poor ionizing solvent, is so much more susceptible to the linear salt effect than acetic acid that with concentrations of lithium perchlorate above 0.036 *N* ether can be made the better ionizing solvent. This effect has been regarded as a special case of electrophilic attack on the leaving group by the ion pair. In certain cases the solvolysis rate deviates sharply downward from the linear curve in the region from zero to a few thousandths normal salt concentration. This "*special salt effect*" (with Fainberg and Robinson, 83, 84, 101–103; Clippinger, 104) was shown to be a form of ion-pair return; however, the special salt effect is not seen in the polarimetric rate constant by which *internal return* is detected. The special salt effect is observed only with respect to the total solvolysis detectable by titrimetric kinetic studies; it indicates the behavior of "solvent-separated," but electrostatically held, ion

(3) P. R. Story and M. Saunders, *J. Amer. Chem. Soc.*, **84**, 4876 (1962).

pairs. Thus the manipulation of these experimental tools was used to distinguish tight ion pairs, solvent-separated ion pairs, and free ions. The last species sometimes play no distinguishable role in solvolysis, but the phenomenon of *common-ion rate depression*, when present, shows that starting material can be reformed also at this third stage of the ionization process (Winstein, Clippinger, Fainberg, Heck, and Robinson, 94).

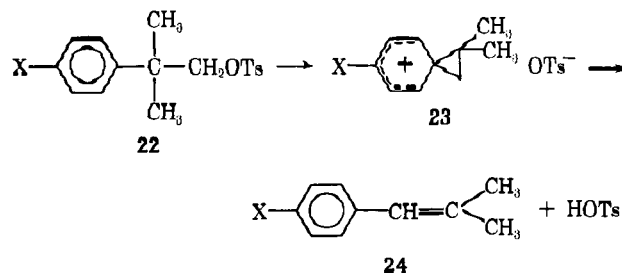
Another phenomenon of ionic behavior discovered by the Winstein group is *merged substitution and elimination* (with Darwish and Holness, 105; Cocivera, 185; Parker, Ruane, and Biale, 257; Biale, Parker, Smith, and Stevens, 281); strong nucleophiles of low proton basicity, once thought capable only of nucleophilic displacement reactions, induce substantial amounts of elimination along with the S_N2 reaction, producing olefins in accord with the Saytzeff rule.

Two widely useful linear free energy relationships were developed in the work on solvolysis. Hydroxylic solvents and solvent mixtures were assigned an ionizing-power parameter Y (Winstein and Grunwald, 56) based on the rate of solvolysis of *tert*-butyl chloride in those solvents. Each ionizable compound could then be assigned a substrate parameter m allowing the correlation of its solvolysis rates in the same solvent series by the equation $\log k/k^0 = mY$, k^0 being the rate constant in 80% ethanol–20% water. The correlations were not perfect for different mixed solvents (Winstein and Fainberg, 100, 112–115); a painstaking investigation of the activation parameters revealed complexities defying analysis.

Winstein, Grunwald, and Jones (64) proposed that *tert*-butyl chloride undergoes solvolysis with essentially no covalent nucleophilic interaction of the solvent with the developing cation, but that for secondary and primary chlorides and sulfonates there is in general a blend of nucleophilic (“ N ”) and pure ionizing (“limiting, Lim ”) action of most solvents. This discussion ignored the mechanism of attack on the anion as being of much less interest for solvolysis, and hence was not forced to consider specific electrophilic activity and susceptibility as in the general treatment of Swain.

More recently, the unraveling of the complex behavior of ion pairs, together with the importance of superacid solutions, revived interest in mechanisms in solvents so little nucleophilic that they might make solvolyses of even primary compounds “limiting.” The reexamination of the most interesting ionic reactions in such solvents as trifluoroacetic and fluorosulfonic acids (274, 275) had barely begun, but the results were spectacular: neopentyl tosylate, which in ethanol solvolyzes 4040 times more slowly than methyl tosylate, is a million times faster than methyl in fluorosulfonic acid at -44° !

Because the ionization (98, 122) of a neophyl tosylate **22** leads to elimination (\rightarrow **24**) instead of displacement by the solvent, the rate of reaction in this case could be used to register the ionizing power of solvents without hydroxyl groups, thus extending such measurements to the whole possible range of solvents (Smith, Fainberg, and Winstein, 157). *p*-Methoxyneophyl tosylate was the substrate of choice, since the ionization occurs exclusively with participation of the aryl group and the ρ^+ parameter is -2.96 (Heck and Winstein, 122).



This work provided a needed comprehensive measure of solvent ionizing power based upon reaction rates. It is parallel in general with the spectroscopically-based Z parameter of Kosower and the E_T of Dimroth, but with some individual transpositions of order.

Unfortunately, this useful parameter lacks a useful name. It is most commonly referred to as “ $\log k_{\text{ion}}$.” The term “ W ,” for Winstein parameter, would be both appropriate and wieldy.

Throughout his scientific life Saul Winstein was driven by a curiosity that could be satisfied only by fundamental understanding. Because current progress in science was always offering the prospect of deeper understanding, he was never really through with a problem, but continued scrutinizing and worrying—to use two of his favorite words—as long as it seemed possible to add to the insights already attained. This possibility was raised whenever a new technique became available—as many did during his career—such as dynamic nmr, reasonable quantum mechanical models of molecular structure, esr, photochemical techniques, improved mass spectrometry, the use of complexation with transition metals, nondestructive superacid solutions, and many others. The practice often recommended to young Ph.D.’s of forgetting about their thesis work and looking elsewhere for a new field would have been unthinkable when, as a new Ph.D., Winstein held some central problems of structure and reaction mechanism in his hand. There are times when the shortest route to the new is a determined attempt to understand the known. Although most of his research projects grew directly out of his experience, they developed rapidly and eventually touched nearly every aspect of rational organic chemistry. In addition to some of the main themes discussed here, Winstein contributed to conformational analysis (with Holness, 92; Lewin, 175), the use of dielectric constant (with Wood, 17; Kumler, Boikess, and Bruck, 192), infrared (with Kivelson, Bruck, and Hansen 162), and nmr (with Anet, Bourn, and Carter, 220, 221; Brookhart and Levy, 242) as structural tools in studying steric compression; preparation and properties of quinone methides (with Filar, 158); charge transfer complexes (with Feldman, 169, 178, 262); Baeyer–Villiger rearrangements (with Hedaya, 177), photoisomerizations (with Zirner, 197), reactions of thermal carbon atoms (with Sprung and Libby, 215), novel reactions of olefin–metal carbonyl complexes (with Kaesz and Kreiter, 224), fluxional systems (*e.g.*, with Childs, 266; Poulter, 271), and oxidation–reduction (with Johnson, 65–67; Traylor, 135; Anderson 182; Rapoport, Sleezer, and Young, 222; Kitching, Rapoport, and Young, 230).

Analyze as we may the chronology and logic of Winstein’s research successes, the question remains how anyone could maintain such a prodigious output

of work of the highest quality and perceptiveness. He was far more than a mere "right man at the right time." Possibly his rarest quality was his total personal identification with the problems of science. He would never forego or curtail an opportunity for scientific discussion. Old friends recall that Winstein's perception foreshadowed some research developments by a number of years, including the homoallylic phenomena worked out in his laboratory, and the importance of conformation, to which the key was later provided by Barton and Hassel. In turn, Winstein's unusually keen awareness of the nature and urgency of the puzzles presented by chemical phenomena provided the incentive for his students to work and excel as they would never do without such leadership.

Winstein wrote as he thought, with emphasis on logic and the search for answers. His papers, being addressed to an imaginary audience of his equals, were clear and free of dramatic or persuasive devices,

although the drama inherent in some of the results is there for all to see.

Winstein's contributions were recognized by many honors. These included, in part, the American Chemical Society Award in Pure Chemistry in 1948 and election to the National Academy of Sciences in 1955. In 1962 he was named California Scientist of the Year, received an honorary doctorate from the University of Montpellier, France, and the Richards Medal of the American Chemical Society. He became a Fellow of the American Academy of Arts and Sciences in 1966, received the Norris Award in Physical Organic Chemistry of the American Chemical Society in 1967, and the Franklin Memorial Award for Outstanding Contributions to Chemistry in 1968. In 1970 he was honored posthumously with the award by President Nixon of the National Medal of Science.

PAUL D. BARTLETT

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3. The Dibromide Method of Analysis of Butene Mixtures. With W. G. Young, *ibid.*, **58**, 102 (1936).

Allylic Rearrangements

4. I. Crotyl and Methylvinylcarbinyl Bromides. With W. G. Young, *ibid.*, **58**, 104 (1936).

5. II. Crotyl and Methylvinylcarbinylmagnesium Bromides. With W. G. Young and A. N. Prater, *ibid.*, **58**, 289 (1936).

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18. Allylic Rearrangements. XIII. Kinetics and Mechanism of Conversion of Butenyl Chlorides to Acetates and Ethers. With W. G. Young and J. D. Roberts, *ibid.*, **64**, 2157 (1942).

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