## Nobel Centennial Essays

# A Century of Chemical Dynamics Traced through the Nobel Prizes

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## 1999: Ahmed H. Zewail

**by J. Van Houten**

### Ahmed H. Zewail (1946– )

*for his studies of the transition states of chemical reactions using femtosecond spectroscopy*

This is the final essay in a series of thirteen accounts, written in commemoration of the centennial of the Nobel Prize, examining the history of chemical dynamics in the 20th century *(1)*. The 1999 chemistry Nobel Prize was awarded to Ahmed Zewail "for his studies of the transition states of chemical reactions using femtosecond spectroscopy." His pioneering investigation of fundamental chemical reactions using ultra-short laser flashes allowed chemists, for the first time, to monitor reactions on the time scale on which the atoms are actually moving as bonds are broken and formed. Achieving this fundamental limit represents the crowning achievement of the century of progress in chemical dynamics that has been discussed in this series.

A previous essay noted *(1i)* that the 1986 Nobel Laureate John Polanyi stated in 1979 that we were then "standing on the threshold of…the last frontier of chemical dynamics, … the spectroscopy of the [transition state]" *(2)*. Near the conclusion of his 1986 Nobel lecture Polanyi spoke of what he called "transition state spectroscopy (TSS)" saying that "attempts are being made to observe the molecular partners while they are, so to speak, on the stage, rather than immediately prior to and following the reactive dance…it is a young but burgeoning field" *(3)*. Polanyi's TSS work represented a time-integrated approach, not anticipating the dream of Zewail's femtochemistry, which resolves the events in time as atoms join in chemical bonding or depart as bonds are broken. In 1995 Polanyi and Zewail co-authored the paper Direct Observation of the Transition State *(4)* in "Holy Grails in Chemistry", a special issue of *Accounts of Chemical Research* dedicated to the memory of Linus Pauling.[1]

### The Development of "Femtochemistry"

Zewail's Nobel lecture opened with a discussion of time measurement traced from the calendars and sundials of the ancient Egyptians to the mechanical clocks of Renaissance Europe to the modern atomic clock *(5)*.[2] He went on to outline the development of chemical dynamics through the 20th century, specifically mentioning contributions by most of the Nobel Laureates who have been covered in this series. He noted how in 1889 Svante Arrhenius conceived the familiar expression for the temperature dependence of the rate constant *(1b)*, which was, itself, rooted in the description of chemical dynamics formulated by Jacobus van't Hoff in 1884 *(1a)*. He included the work on catalysis by Wilhelm Ostwald in the "arrow of time" used to depict progress in chemists' ability to study increasingly fast reactions *(1c)*. He described the studies of gas phase chemical kinetics in the 1920s that led Cyril Hinshelwood *(1e)* and others to develop elementary mechanisms for unimolecular reactions having different steps describing activation, energy redistribution, and chemical rates.


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Moving on to a description of developments in the middle of the 20th century, Zewail related contributions by Rudy Marcus that brought into focus the nature of initial and transition state vibrational modes (RRKM theory), and also the Marcus theory of electron transfer in solution *(1j)*. He acknowledged the significant stride into the microsecond regime resulting from the development of flash photolysis by Ronald Norrish and George Porter and of temperature- and pressure-jump methods by Manfred Eigen *(1f)*; noting that these rapid techniques allowed short-lived intermediates to be observed for the first time. Zewail noted in his Nobel lecture *(5)* that Eigen had titled his 1967 Nobel lecture "Immeasurably Fast Reactions", going on to say that Eigen "told me, when I teased him about his title that nobody in the 1950s anticipated the laser and the short pulses they can provide." As the century progressed, lasers replaced flash-lamps, and the duration of the pulses was reduced into the nanosecond, picosecond and, ultimately, the femtosecond regime. In the introduction to the first volume of a series on femtosecond chemistry, George Porter wrote in 1995: "The study of chemical events that occur on the femtosecond time scale is the ultimate achievement in half a century of development and, although future events will be run over the same course, chemists are near the end of the race against time" *(6)*. Zewail asserted that moving from the microsecond regime of Eigen, Norrish, and Porter into the femtosecond regime "opened the door [as] the transition state, the cornerstone of reactivity, could be clocked as a molecular species $TS^{\ddagger}$, providing a real foundation to the hypothesis of Arrhenius, Eyring, and [Michael] Polanyi for ephemeral species $[TS]^{\ddagger}$ and leading the way to numerous new studies" *(5)*. He continued, "the previous virtual status of the transition state has now given way to experimental reality" and that "inferences deduced from 'rotational periods' as clocks can now be replaced by the actual clocking of nuclear (vibrational) motion."

The magnitude of the femtosecond can be appreciated in several ways. The vibrating atoms in a molecule typically move at a speed of approximately 1 km/s, so an atom requires 100 fs to move a distance of 1 Å. Light moving at $3 \times 10^8$ m/s travels only 30 μm during that 100 fs. Eigen, Norrish, and Porter extended chemists' ability to study reactions into the sub-millisecond regime *(1e);* the ratio of a femtosecond to a millisecond equals the ratio of a millisecond to the 32 years that elapsed between the Nobel Prizes in 1967 and 1999.

The femtosecond laser has sometimes been characterized as an ultra-fast descendent of the flash lamp. However, it is much more than that, and Zewail has emphasized that coherence is as important as speed in femtochemistry. Professor Bengt Nordén, member of the Nobel Committee for Chemistry, described coherence as "the crucial point of Zewail's femtochemistry: to use a femtosecond pulse to, so-to-say, blow the whistle to start all the molecules at the same time and at the same point of their vibration cycles. A consequence of the synchronization of the molecules ('coherent preparation' of the system), is that although the measurement by the laser probe pulse will include a huge number of molecules, typically millions, their behavior will not have blurred out but, thanks to the coherence, be like that of a classical vibrator, like a vibrating spring. In turn this enables observation of the movements of the nuclei during the vibration and thus a characterization of the transition state with high spatial resolution (about 0.1 Å in NaI [one of the first systems Zewail studied])." *(7).* In his Nobel lecture *(5)* Zewail explained that with femtochemistry "the ephemeral transition states, denoted in the past by square brackets ($[TS]^{\ddagger}$) for their elusiveness, can now be clocked as molecular species $TS^{\ddagger}$. Moreover, on this time scale, the time-dependent description of a coherent single-molecule trajectory represents the classical nuclear 'motion picture' of a reaction as its wave packet proceeds from the initial state, through transition states, and on to the final products—the language of actual dynamics! The femtosecond time scale is unique for the creation of such coherent matter waves on the atomic scale of length, a basic problem rooted in the development of quantum mechanics and the duality of matter."

When Nordén introduced Zewail at the Nobel award ceremony, he said: "Zewail's use of the fast laser technique can be likened to Galileo's use of the telescope, which he directed towards everything that lit up the vault of heaven. Zewail tried his femtoscope laser on literally everything that moved in the world of molecules. He turned his telescope towards the frontiers of science" *(8a).* Zewail began by studying relatively simple systems in the early 1980s and, during the ensuing decades, he has expanded his investigations to a diverse array of systems in all branches of chemistry as well as biology and medicine.

Zewail's first experiment with sub-picosecond resolution involved the unimolecular decomposition of iodine cyanide into an iodine atom and a cyano radical: ICN → I + CN. In 1985 Zewail's group succeeded in observing the separation of the CN radical product on a 500 fs time scale, but he realized that a new laser system was necessary to achieve femtosecond resolution. In 1986 the new femtosecond laser system was installed in the Caltech laboratory that had formerly housed Linus Pauling's X-ray apparatus. Using that system, Zewail was able to observe the transitory species $I\cdots CN^{*\ddagger}$. Zewail said of this work: "The first $I\cdots CN^{*\ddagger}$ transient surprised us, but after long and late hours of discussions and control experiments it became clear that, indeed, the transition configurations and the final products could be monitored in real time. We submitted our first communication to the *Journal of Chemical Physics* (received June 3, 1987), and it was accepted on June 15, 1987 *[9].* The referee of this report was not only prompt, but also, in retrospect, visionary. His report was ultrashort: 'It [the manuscript] has the smell that the authors are onto some very exciting new stuff…It may turn out to be a classic. Publish with all dispatch.'"(5). Nevertheless, the title of that paper, "Real-time femtosecond probing of 'transition states' in chemical reactions," was characterized as "somewhat sensational" in the Nobel press release announcing Zewail's 1999 award (*7;* see the "extended version in English").

The importance of coherence was dramatically illustrated in Zewail's next series of experiments—the decomposition of alkali halides, in particular NaI. He said: "The reactions of the alkali halides were thought of as perfect prototypes. Since they involve two potentials (covalent and ionic) along the reaction coordinate, I thought we would have fun with these systems. Moreover their historical position in crossed molecular beam experiments [of the 1986 Nobel Laureates, Dudley Herschbach and Yuan Lee *(1i)*] (the 'alkali age') made them good candidates for the 'femto age.' The reaction of NaI, unlike ICN, involves two electronic coordinates and one nuclear coordinate, the separation between Na and I" *(5).* In that experiment, published in 1988, "bursts" of free sodium atom fragments were found to correlate with oscillations between the covalent and the ionic forms of NaI as a result of the vibrational motion of the NaI *(10).* Zewail said: "The results also illustrated the importance of coherent wave packets in quasi-bound systems. The NaI experiment was a watershed event leading to an entirely new paradigm in the field of femtochemistry and establishing some new concepts for the dynamics of the chemical bond…this is the world of dynamics *not* kinetics" *(5).*

Zewail's femtochemistry work has intersected with that of all twelve of the previous Nobel Prizes in chemical dynamics discussed in this series, seven of which have already been mentioned above *(1).* Zewail's investigation of the decomposition of cyclobutane to form ethylene (or the reverse—formation of cyclobutane from two ethylene molecules) is a classic test of the symmetry rules for which Roald Hoffmann received the 1981 Nobel Prize *(1g).* His investigations involving transition metal electron transfer reactions utilize some of the same complexes first studied by Henry Taube, the 1983 Nobel Laureate *(1h).* He has investigated reactions of simple halocarbon molecules that are very similar to those identified by the 1995 Nobel Laureates, Paul Crutzen, Mario Molina, and Sherwood Rowland, as being responsible for stratospheric ozone depletion *(1j).* He has utilized ab initio and density functional theory methods developed by the 1998 Nobel Laureates, John Pople and

Walter Kohn, to model some of his experimental dynamics results *(1l)*. The only exception to the intersection between femtochemistry and the twelve previous Nobel Prizes in chemical dynamics is George de Hevesy's 1943 award for isotope tracers *(1d)*. In a sense, this is the proverbial "exception that proves the rule" inasmuch as femtochemistry looks at the intimate details of the transition state whereas the isotope tracer technique provides information on the overall net reaction mechanism. Nevertheless, Zewail has used isotopes to study the dynamics of proton transfer in chemical and biological systems *(5)*.

The limitations of this essay preclude anything approaching an adequate discussion of the fascinating subject of femtochemistry and the myriad systems that Zewail has characterized as "Femtocopia" *(5)*. Fortunately, there are a number of reports available that cover this subject, and the interested reader is encouraged to consult the excellent articles listed in Further Reading and Literature Cited.

## Zewail's Background

Ahmed Zewail was born in the Nile River delta town of Damanhur, and raised in the nearby town of Disuq, about 60 km south of the Egyptian city of Alexandria. He is the first person from the Arab world to receive a Nobel Prize in science.[3] Zewail's father ran a small business importing and assembling bicycles and motorcycles; later he obtained a government civil service post. His extended family is concentrated in the area around Damanhur and Alexandria, where they hold influential positions as university professors, judges, and business leaders. Zewail's childhood and adolescence in Disuq were steeped in the culture of Egypt and the Islamic religion; for him the local mosque was a center of culture and scholarship as well as religion. He has retained a strong affiliation with his family and his boyhood home.

The local primary and secondary schools provided Zewail with a well-rounded education. He has shown his gratitude by donating a portion of his Nobel award to fund equipment and infrastructure at the Disuq high school and to provide a prize honoring the top graduate of the school, which is now named for him *(8b)*. As a young student he excelled in chemistry, physics, and mathematics, but he also received a strong background in subjects such as Arabic and history and he enjoyed participating in activities such as photography, art, music, and theater. He constructed his own chemical apparatus from an Arabic coffee maker, and performed some of his earliest experiments in chemical dynamics when he ignited the gas formed upon heating some wood in a sealed tube.

High scores on his secondary school science and math examinations gained Zewail an appointment as an undergraduate student in the Faculty of Science at the prestigious Alexandria University. In contrast to the American educational system, Egyptian students are expected to enter the university with a strong background in the general "liberal studies" areas; at the university they concentrate only on subjects related to their major. Thus Zewail enrolled in chemistry, physics, mathematics, and geology in his first year and, in keeping with the Egyptian university curriculum, he narrowed the focus of his studies first to physics and chemistry and then just to chemistry as he progressed through his four undergraduate years.

As a result of his excellent scholastic achievement in his first two years at Alexandria University, Zewail was selected by the faculty as one of a small group for special study. They were known by the English word "special," a designation similar to what we would call an honors student. Zewail said of this appointment: "I felt my horizons were just beginning to expand, because it was known that specials would become *mu'ids* ('demonstrators', equivalent to a graduate teaching/research assistant) and then, if you became a *mu'id*, you would go on to become a university professor after obtaining a Ph.D. So making 'special' was a big thing" *(8c)*.

After completing his B.S. at the top of his class in the sciences in 1967, Zewail was appointed as a *mu'id* at Alexandria University. Although the normal duties of a *mu'id* were similar to those of a graduate laboratory teaching assistant in the U.S., Zewail was asked to conduct evening review sessions for a class of about five hundred students in general chemistry. He said that the students attending his reviews would fill the lecture hall because he "had already acquired a reputation for a simplicity and clarity as a lecturer" *(8d)*. In words reminiscent of those of other Nobel Laureates discussed in this series—Roald Hoffmann *(1g)*, Henry Taube *(1h)*, and Mario Molina *(1k)*—Zewail said regarding his early teaching experience: "To this day, this is part of the enjoyment I experience in simplifying complicated concepts. I believe that behind every important and fundamental concept there must be simplicity and clarity of thought. If it's fuzzy and unclear, and one is making it complicated, then I'm not sure we have an understanding of it yet."

Zewail completed his spectroscopic research and wrote his M.S. thesis in only eight months. His graduate mentors, both junior faculty members who had received their Ph.D.s in the U.S., encouraged him to go abroad to complete his graduate work. He was accepted into the Ph.D. program at the University of Pennsylvania in April 1969. However, the world political situation and some bureaucratic problems stood in the way of his departure from Egypt (Egypt had been at war with Israel and was most closely aligned with the Soviet Union), so Zewail had to make a number of trips to Cairo to obtain the required permission to leave the country for the U.S. He took a leave of absence from his position at Alexandria University and left for Philadelphia in August 1969 with $40, the maximum amount he was permitted to take with him *(8e)*.

When Zewail arrived in Philadelphia, he spoke little English and had some interesting experiences becoming adjusted to American ways. However, he quickly settled into the scientific work in Robin Hochstrasser's lab at Penn, where he found the atmosphere very stimulating. His first project, on the Zeeman effect of the triplet state in sodium nitrite, resulted in a publication within a year. Three more papers on the Zeeman effect (in benzene and triazene) were published the following year. He reported: "The diverse research problems I worked on, and the collaborations with many able scientists, were both enjoyable and profitable…I was learning new things literally every day. The atmosphere…was most stimulating and I was enthusiastic about researching in areas that crossed the disciplines of physics and chemistry (sometimes too enthusiastic!)….I was working almost 'day and night' and doing several projects at the same time: the Stark effect of simple molecules; the Zeeman effect of solids like $NO_2^-$ and benzene; the optical detection of magnetic resonance (ODMR); double resonance techniques, etc. Now, thinking about it, I cannot imagine doing all of this again, but of course then I was 'young and innocent'" *(7).* By the time Zewail completed his Ph.D. in 1973 he had twelve publications to his credit *(8f)*.

Zewail reported that he "had strong feelings about returning to Egypt to be a university professor, even though at the beginning of my years in America my memories of the frustrating bureaucracy encountered at the time of my departure were still vivid. With time, things change, and I recollected all the wonderful years of my childhood and the opportunities Egypt had provided to me. Returning was important to me, but I also knew that Egypt would not be able to provide the scientific atmosphere I had enjoyed in the U.S. A few more years in America would give me and my family two opportunities: first, I could think about another area of research in a different place (while learning to be professorial!). Second, my salary would be higher than that of a graduate student, and we could then buy a big American car that would be so impressive for the new professor at Alexandria University! I applied for five [postdoctoral] positions, three in the U.S., one in Germany, and one in Holland, and all of them with world-renowned professors. I received five offers and decided on Berkeley" *(7).*

Zewail reported that his postdoctoral experience working with Charles Harris at Berkeley "was an important transition in my scientific career, because it set me on a course of research that…shaped [my scientific future]" *(8g).* His experience with coherence in spin systems for excitation transfer of interacting dimers allowed him to become familiar with the theory of coherence and to study the fundamentals of the related fields of quantum optics and quantum electronics. His first independently authored paper came as he extended the concept of coherence to multidimensional systems. Zewail received an IBM fellowship that allowed him to spend a second year at Berkeley. He described how his future research with fast kinetic techniques was forged when "Charles decided to build a picosecond laser, and two of us in the group were involved in this hard and 'non-profitable' direction of research(!); I learned a great deal about the principles of lasers and their physics" *(7).*

As he embarked on a search for his first faculty position in 1976, Zewail had the option of returning to the position that was being held for him at the Alexandria University. However, after being encouraged by his mentors to apply for positions at U.S. universities, he received offers from several major research institutions. After considering his options carefully, he decided to accept a position at the California Institute of Technology, explaining that he was attracted to Caltech because he was convinced it would provide the most supportive research environment for a beginning researcher in his field *(8h)*. It is apparent that he was correct in that assessment: he rose quickly through the ranks and has remained at Caltech throughout his career. He was named the Linus Pauling Professor of Chemical Physics in 1990 and, in 1995, the Linus Pauling Chair Professor of Chemistry and Professor of Chemical Physics. In 1996 he was appointed as Director of the NSF Laboratory for Molecular Sciences at Caltech, which has given him the opportunity to be directly involved in myriad research projects.

## Notes

1. Linus Pauling is the only person to receive two unshared Nobel Prizes—the Chemistry Prize in 1954 and the Peace Prize in 1962. He died in 1994 at the age of 93, a few months before the publication of the March 1995 issue of *Accounts of Chemical Research* on "Holy Grails in Chemistry" that was dedicated to his memory.

2. Norman Ramsey shared the physics Nobel Prize in 1989, in part, for developing the cesium atomic clock that is now the recognized scientific fundamental standard for time.

3. Prior to Zewail, there were two Egyptian Nobel Laureates—the novelist Naguib Mahfouz received the Literature Prize in 1988, and President Mohamed Anwar al-Sadat shared the 1978 Peace Prize with Menachem Begin. Strictly speaking the first Egyptian-born science Laureate was Dorothy Crowfoot Hodgkin, who received the chemistry Nobel Prize in 1964. Although Hodgkin was born in Cairo in 1910, the daughter of a British archaeologist then stationed in Egypt, she was a British subject and spent most of her formative years as well as her entire professional career in England.

## Further Reading


Zewail's Nobel lecture and his published autobiography appear in refs. *5* and *8*.

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The Femtoland Web page is at *http://www.its.caltech.edu/~femto* (accessed Sep 2002).

### [W]Supplemental Material

A list of all recipients of the Nobel Prize in Chemistry, their affiliations, and the work for which the award was made, is available in this issue of *JCE Online*.

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## Nobel Centennial Essays

### Series Conclusion

The essays in this series have traced the development of chemical dynamics through 13 Nobel Prizes awarded to 22 individuals during the 20th century. Professor Bengt Nordén concluded his 1999 Nobel presentation address by saying: "Femtochemistry has radically changed the way we look at chemical reactions. A hundred years of mist surrounding the transition state has cleared. …[Zewail's] pioneering work has fundamentally changed the way scientists view chemical reactions. From being restricted to describe them only in terms of a metaphor, the transition state, we can now study the actual movements of atoms in molecules. We can speak of them in time and space the same way we imagine them. They are no longer invisible" *(8i).* Thus the Nobel Prize awarded to Ahmed Zewail in 1999 for developing femtochemistry serves as a fitting climax to a century of progress in chemical dynamics.

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