# SEVENTEENTH ANNUAL CHARLES A. COULSON LECTURE

**Tuesday, April 26, 2004**
**11:00 A.M. Room 400, Chemistry**

**Sason Shaik**

" Is There Anything New in Chemical Bonding?
New Bonding Concepts from Valence Bond Theory"

**Presented by**

**Center for Computational Chemistry**

**University of Georgia**

## CHARLES A. COULSON

Charles Coulson was born on December 13, 1910 in Dudley, England, the elder of twin sons. His forebears had been farmers, but his father was then the Principal of the local Technical College. It was his father who modeled for him the Christian faith which throughout his life was to make every scientific achievement an act of service.

In 1920 the family moved to Bristol on the appointment of Alfred Coulson as H. M. Inspector of Technical Colleges for South West England, and in 1923 Charles was awarded a scholarship to Clifton College. In 1928 he entered Trinity College, Cambridge, and after taking First-class Honors in the Mathematical Tripos in 1931, he took the unusual step of staying on a further year to study physics and took First-class Honors in the Natural Science Tripos. He was elected to a Fellowship of Trinity College and was attracted by the blossoming field of wave mechanics, heralded as the key to an understanding of chemistry. He received his Ph.D. in 1935 working with Professor John Lennard-Jones. His thesis work - completed more than sixty years ago - included perhaps the first *ab initio* calculation on a polyatomic molecule, the ion $H_3^+$.

During the decade 1936-46 he laid the foundation for many developments in theoretical chemistry, producing more than forty significant papers. After his marriage in 1938 he moved to Dundee. In 1941 he was elected a Fellow of the Royal Society of Edinburgh. In 1945 he moved to Oxford as an ICI Fellow and in 1947 to London to the Chair of Theoretical Physics at King's College, where he built up a new department and greatly expanded his research group. He was prolific in the application of simple MO theory to problems in organic chemistry; particularly to large condensed ring systems, to theories of reactivity and carcinogenesis, to strained molecules, and to free radicals. But along with David Craig and others, he was also among the pioneers in the systematic use of configuration interaction; and with Roy McWeeny he completed work on the direct calculation of wave functions in momentum space. At this time he also broke into new fields; long range forces between large molecules; the solid state, with band theory calculations on graphite and related compounds; and with some of the earliest work on surface states - now so important in theories of catalysis.

He was elected to Fellowship of the Royal Society in 1950 and in the years that followed was the recipient of many prizes and distinctions. In 1951 he returned to Oxford, to the Rouse Ball Chair of Mathematics and to a Fellowship at Wadhaven College, leaving behind in London an established and flourishing Department of Theoretical Physics. In 1972 Charles Coulson became Oxford's first Professor of Theoretical Chemistry.

During most of his work from 1945 onwards, Coulson's scientific activity was concerned more with the building of simplified theories than with direct attempts at solution of the many-electron Schrödinger equation. The beautiful series of papers "The Electronic Structure of Conjugated Systems", written in collaboration with Longuet-Higgins, is the epitome of this philosophy; the model to be used is carefully defined and all its implications are then worked out with rigor and elegance. Most of the chemically important concepts developed in these papers have, in fact, survived the passage of time: the charges and bond orders are the elements of density matrices; the atom and bond polarizabilities are related to response functions; and the whole mathematical framework is strong enough to allow a complete generalization from the Hückel model to *ab initio* many-electron theory without significant modification of the underlying concepts.

From the early fifties Charles Coulson was probably as much in demand as a lecturer on spiritual and ethical issues as he was in scientific circles. In 1959 he became Vice President of the British Methodist Conference, and during his year of office delivered well over 100 lectures or sermons.

**SASON SHAIK**

Sason Shaik was born in Baghdad, Iraq on December 17, 1948. His family immigrated to Israel in the Exodus of the Iraqi Jewry in 1951. He studied chemistry and received his B.Sc. and M.Sc. degrees from Bar-Ilan University. In 1974 he began his Ph.D. studies at the University of Washington under Nicholaos Epiotis. In 1978/9 he spent a postdoctoral year with Roald Hoffmann at Cornell University. He started his first academic position as a Lecturer at Ben-Gurion University, where he became Professor in 1988. In 1992 he moved to the Hebrew University, where he is currently Professor of Chemistry and Director of the Lise Meitner-Minerva Center for Computational Quantum Chemistry.

Sason Shaik has been a Fulbright Fellow (1974-1978), received the Israel Chemical Society Prize for the Outstanding Young Researcher (1987), the Bergman Prize given by the Israeli Academy of Sciences (1995), the Lise Meitner-Alexander von Humboldt Senior Award (1996-1999), the Israel Chemical Society Prize (2001), the Kolthoff Prize (2001), and was elected a Fellow of the American Association for the Advancement of Science in 2004. He has presented several named lectureships and invited series of lectures; most recently (2004) he was named the Kurt Alder Lecturer at the Department of Chemistry, University of Cologne. He appeared in the ISI list of 1000 most cited chemists during 1981-1997.

Professor Shaik's research interests are in the use of quantum chemistry, and in particular of valence bond (VB) theory, to develop concepts that can systematize data and lead to the generation of new problems. In 1981, he published a seminal work that describes the origins of barrier formation in any chemical reaction. This work was followed by numerous publications, some with coworkers Pross, Hiberty, and Wu. This research showed the applicability of the VB model to chemical reactions, starting from classical physical organic reactions, and extending to DNA repair mechanisms, metallo-organic species, and enzymes. To date Shaik's VB diagram model is one of the most complete qualitative models of chemical reactivity, with both predictive and organizing capabilities, as well as a rigorous quantitative foundation. The model has been incorporated into textbooks (e.g., Lowry and Richardson, *Mechanism and Theory in Organic Chemistry*) and monographs (e.g., Eberson, *Electron Transfer Reactions in Organic Chemistry*), alongside the Woodward-Hoffmann Rules and the Marcus Model.

In an important 1984 paper, Shaik proposed the distortive behavior of p-electrons as a general feature of deloclalized species. In the extensive research that followed, Shaik and Hiberty quantitatively established the notion of p-distortivity and showed its link to experimental observables (e.g., structures and IR spectra of excited states of benzene). Two additional important achievements of this work were: (i) a derivation of a general model of electronic delocalization in isoelectronic families, covering the range of stable delocalized species, Psuedo-Jahn-Teller unstable species, and Jahn-Teller distorted species; (ii) the creation of a portable VB theory of delocalization free from the traditional failures of resonance theory. This research has also been incorporated into monographs and textbooks. In 1994, Shaik derived the concept of two-state reactivity (TSR) in collaboration with Helmut Schwarz. The TSR concept resolved some intriguing findings in C-H activation by transition metal-oxo cations and has turned out to be a general paradigm for catalytic bond activation in organometallic chemistry. Since 1998, Shaik and his coworkers have extended the TSR concept to cytochrome P450 and other heme enzymes, resolved a major controversy in the area of hydroxylation by P450, and demonstrated the wide applicability of the mechanism to oxidative processes.

Most recently, Professor Shaik and coworkers showed that the active species of cytochrome P450 enzymes, so-called Compound I, behaves as a chameleon species that adapts its geometry and electronic structure to the specific protein environment. Thus, the use of a single reagent of a variable nature, susceptible to the environment, is an effective strategy by which the enzyme modulates its selectivity. The chameleon oxidant concept should have wide ranging implications for the field of enzymatic catalysis.

Beginning in 1991, Sason Shaik has been engaged in the furtherance of chemical education, targeting the social sciences and humanities. This activity has led him to give popular talks about chemistry. His recent popular essay entitled, "Chemistry- A Central Pillar of Human Culture", reflects Professor Shaik's continuing fascination with chemistry.

*"I want to stress the power of a modern computer. In favorable cases its final numerical values have an accuracy as great as or even greater than, that of the best experiment. The computer is a tool, just as is a spectroscope or a calorimeter. Indeed a recent report by the National Academy of Sciences in the U.S.A. concluded that 'quantum chemistry can now be regarded as a highly refined instrument on par with, or even superior to, the finest laboratory instruments'. But this does not mean it should always be used--the laboratory instruments will often be more appropriate. And if we use a computer without a clear idea of why we do it, we are really wasting time and money."*

**Charles A. Coulson**
**Inaugural Lecture as**
**First Professor of Theoretical Chemistry**
**The University of Oxford**
**February 13, 1973**

**Named Lectures at the Center for Computational Quantum Chemistry**

**Robert S. Mulliken**

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|---|---|
| 1988 | Robert G. Parr |
| 1989 | Clemens C. J. Roothaan |
| 1989 | Klaus Ruedenberg |
| 1990 | Michael Kasha |
| 1990 | Harrison Shull |
| 1991 | A. Douglas McLean |
| 1991 | Megumu Yoshimine |
| 1992 | Enrico Clementi |
| 1992 | Ernest R. Davidson |
| 1993 | Reinhart Ahlrichs |
| 1993 | Rodney J. Bartlett |
| 1993 | Bowen Liu |
| 1994 | Alexander Delgarno |
| 1994 | David R. Yarkony |
| 1995 | Charles W. Bauschlicher, Jr. |
| 1995 | Martin Quack |
| 1995 | Leo Radom |
| 1996 | Paul S. Bagus |
| 1996 | Sigrid D. Peyerimhoff |
| 1997 | Werner Kutzelnigg |
| 1998 | Roald Hoffmann |
| 1998 | Peter R. Taylor |
| 1998 | Dieter Cremer |
| 1999 | Gustavo E. Scuseria |
| 1999 | Timothy J. Lee |
| 2000 | Wilfried Meyer |
| 2000 | Axel Becke |
| 2001 | Russell M. Pitzer |
| 2001 | Dudley R. Herschbach |
| 2002 | Yuan T. Lee |
| 2002 | William A. Lester |
| 2002 | Stephen R. Leone |
| 2002 | John F. Stanton |
| 2003 | Jack Simons |
| 2003 | Donald G. Truhlar |
| 2003 | Krishnan Balasubramanian |
| 2004 | Richard A. Friesner |
| 2004 | Eluvathingal D. Jemmis |
| 2005 | F. Albert Cotton |

**Charles A. Coulson**

| | |
|---|---|
| 1988 | Michael J. S. Dewar |
| 1989 | John A. Pople |
| 1990 | Carl M. Moser |
| 1990 | Isaiah Shavitt |
| 1991 | William A. Goddard III |
| 1991 | Nicholas C. Handy |
| 1991 | Norman H. March |
| 1992 | Peter Pulay |
| 1992 | Bjorn O. Roos |
| 1993 | Keiji Morokuma |
| 1993 | Per E. M. Siegbahn |
| 1994 | Jan E. Almöf |
| 1994 | Clifford E. Dykstra |
| 1994 | Mostafa A. El-Sayed |
| 1995 | William H. Miller |
| 1995 | Paul R. Schleyer |
| 1996 | Martin Karplus |
| 1996 | Hans-Joachim Werner |
| 1996 | Barbara J. Garrison |
| 1997 | Thom H. Dunning |
| 1997 | Mark S. Gordon |
| 1998 | Krishnan Raghavachari |
| 1998 | Walter Thiel |
| 1998 | A. David Buckingham |
| 1999 | W. Graham Richards |
| 1999 | Trygve Helgaker |
| 2000 | Frank A. Weinhold |
| 2000 | Michael J. Frisch |
| 2001 | Petr Carsky |
| 2001 | C. Bradley Moore |
| 2001 | Ivan Hubac |
| 2002 | Yitzhak Apeloig |
| 2002 | Yngve Öhrn |
| 2002 | Martin Head-Gordon |
| 2003 | Ahmed Zewail |
| 2003 | Helmut Schwarz |
| 2004 | Josef Paldus |
| 2004 | Gernot Frenking |
| 2005 | Sason Shaik |