

Josef Paldus

1935 - 2022

an Obituary by Josef Michl, Petr Čársky and Miroslav Urban

Josef Michl: Josef Paldus, 87, passed away on November 15, 2022, in Waterloo, Ontario, Canada. He was born on November 25, 1935, in Bzí near Železný Brod in what was then Czechoslovakia and now is the Czech Republic, obtained his undergraduate education in the Department of Mathematics and Physics at the Charles University, and his doctorate with Prof. Jaroslav Koutecký at the Institute of Physical Chemistry of the Czechoslovak Academy of Sciences, both in Prague. His PhD dissertation dealt with the first interpretation of electronic spectra of pi-electron systems with transannular interactions. He subsequently spent two postdoctoral years in the laboratory of D. A. Ramsay at the National Research Council of Canada, investigating fine structure in the spectra of glyoxal. After return to the Institute of Physical Chemistry in Prague he started an independent career in quantum chemistry that was interrupted by the Russian invasion of the country in August 1968. Like many others, he and Eva, his wife and life-long companion, decided to emigrate. Joe accepted a faculty position in the Department of Applied Mathematics at the University of Waterloo in Ontario, Canada, where he remained active for the rest of his life. Their daughter Barbara, fluent in five languages, obtained her doctorate at Stanford University, and has developed a very successful career in business.

Joe has made several very substantial contributions to the development of quantum chemistry. He himself considered as most important his development of unitary group methods for the treatment of correlation energy, permitting an extension of configuration interaction methods to billions of configurations. However, also his collaborative work with a friend and colleague, Jiří Čížek, on Hartree-Fock instabilities and particularly, on the development of coupled cluster methods, has had great impact. Their 1972 paper with Shai Shavitt is viewed as the first ab initio calculation by the CCSDT method, the gold standard today.

I relied on Joe frequently when I was faced with mathematical problems and he always provided valuable insight. His impact on others is best described in the personal reminiscences written by two of his disciples and closest friends, Petr Čársky and Miroslav Urban.

Petr Čársky: I became acquainted with Joe Paldus in the early sixties as a graduate student of Rudolf Zahradník, a colleague of Joe's at the Institute of Physical Chemistry of the Czechoslovak Academy of Sciences in Prague. I was thus able to witness the development of coupled cluster theory in its prenatal and baby stages. The working equations were derived by Jiří Čížek, a student of Jaroslav Koutecký. He published them in a highly mathematical paper [J. Čížek: *J. Chem. Phys.* 45, 4256 (1966)] that attracted little attention for a considerable time. Koch and Kutzelnigg's comment was typical: "Čížek's paper was hard to read, and this not only because it was formulated in terms of diagrams at a time when diagrammatic techniques were not yet as popular in theoretical chemistry as they are nowadays" [S. Koch and W. Kutzelnigg: *Theor. Chim. Acta* 59, 387 (1981)]. The equations deduced from the diagrams did not look friendly for computer coding at that time, either.

Further development of coupled cluster theory was accelerated when Joe Paldus joined the project. Unlike Jiří, Joe was interested in validation of theory by computations. However, the computational facilities available in the country were inadequate and their calculations were limited to semiempirical models, which were unable to fully prove the merits and power of their method. The social and general life conditions under the communist regime were highly unsatisfactory and the Russian invasion of August 1968 finally prompted the families of Čížek and Paldus to emigrate. Their collaboration continued in the Department of Applied

Mathematics at the University of Waterloo in Ontario, Canada. The situation is well characterized by a lament “This work has been initiated in the Institute of Physical Chemistry of the Czechoslovak Academy of Sciences and was interrupted by the presence of foreign armies in the country” [J. Paldus: *Theory and Applications in Computational Chemistry: The First Fifty Years*. Chapter 7. Elsevier 2005]. The same article however also states “Soon after our arrival to Waterloo Jiří and myself were fortunate to make true personal acquaintance of Isaiah (Shai) Shavitt, whose work we knew well from the literature”. Shavitt became interested in the coupled cluster model and this resulted in a joint publication that extends Čížek’s original concept by introduction of singly and triply excited configurations and uses a true ab initio approach [J. Paldus, J. Cizek, I. Shavitt: *Phys. Rev. A*5, 50 (1972)]. In retrospect, in spite of the use of some justified approximations, their extended model may be regarded as a predecessor to what is nowadays called CCSDT, and the paper is a milestone in quantum chemistry. Along with others, Čížek and Paldus contributed greatly to widespread use of coupled cluster theory by elucidating its relation to perturbation theory and configuration interaction, thus making it easily understandable to a wide community of theoretical chemists. After Čížek redirected his interest to other theoretical problems, Paldus became famous by developing new approaches combining the merits of coupled cluster theory and configuration interaction. Remarkable is his reformulation of configuration interaction in terms of the Graphical Unitary Group Approach (GUGA), where what is now called the Paldus tableau facilitates the calculation greatly.

After Joe’s emigration I lost contact with him. Shortly after the Russian occupation a tough communist regime was reinstalled, emigrants were called traitors, and members of the Czech academic community were instructed not to have any contact with them. Toward the end of the seventies the regime became somewhat less oppressive and I was allowed to make short trips to the West. It was a great pleasure and joy for me to meet Joe at conferences abroad. After the fall of communism in 1989 our meetings became more frequent and I was happy that our encounters gradually transformed our acquaintance to a warm friendship that enriched my life.

Miroslav Urban: I visited Josef Paldus at the Institute of Physical Chemistry of the Czechoslovak Academy of Science in Prague repeatedly in the autumn of 1967 and 1968. In 1967 I was accepted as an external PhD student of Professor Jaroslav Koutecký and Josef became my tutor. At that time Josef’s main scientific interest was changing from spectroscopy to many-body theories in quantum chemistry. I was really impressed by his explanation of all aspects of perturbation theory. I could ask anything and his responses were always deep, clear, and understandable. During this period we did not talk about coupled cluster theories, but I lived for years from the background given to me by Joe. It was not just perturbation theory, but also chemistry, particularly spectroscopy. His broad knowledge was unbelievable.

In mid-sixties, quantum chemistry was still dominated by semiempirical methods. Joe realized that the full acceptance of many-body theories should be based on ab initio calculations. I was given a listing of the Polyatom 1 ab initio program and he asked me to implement it as a part of my thesis. The first version of this program did not use contracted Gaussian basis sets, so the first step was to implement this technique. The idea was clear: based on an ab initio Hartree-Fock reference, to proceed with many-body perturbation theory to treat electron correlation properly. Without stating it explicitly, the next step would have been the Coupled Cluster theory, but all plans were interrupted by the Russian invasion of August 1968 and the following occupation. Josef Paldus, Jiří Čížek, Jaroslav Koutecký, Josef Michl, and other prominent scientists left the country. Any contact with “refugees”, in writing or by phone, was considered

by the communist authorities as illegal. Nevertheless, the inspiration from Joe and other celebrities from the Prague Institute of Physical Chemistry given to the quantum chemistry community in Bratislava lasted long and can hardly be overestimated. It led to our own development of ab initio methods including full fourth order perturbation and coupled cluster programs and to numerous applications. Collaboration with those friends from the Prague school established by Josef and Rudolf Zahradník who stayed in Prague after 1968 continued.

New opportunities opened after the “velvet” revolution of 1989. After years, Josef now could visit Prague and Bratislava, and we enjoyed discussions with him at conferences and on other occasions. I can hardly forget Eva’s and Joe’s great hospitality during my stay in Waterloo in 2005. Afterward, we published jointly with Josef three papers on Hartree-Fock stability and broken symmetry solutions of anions in the external confinement followed by Coupled Cluster calculations of properties of confined systems, including in-crystal polarizabilities of a series of inorganic anions. The topic of stability conditions and the broken symmetry problem in quantum chemistry has been investigated by Josef Paldus and Jiří Čížek since 1967 in a series of highly cited papers and remains topical to this day, but it remains in the shadow of their great achievements in the developments of many-body theories.