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Foreword

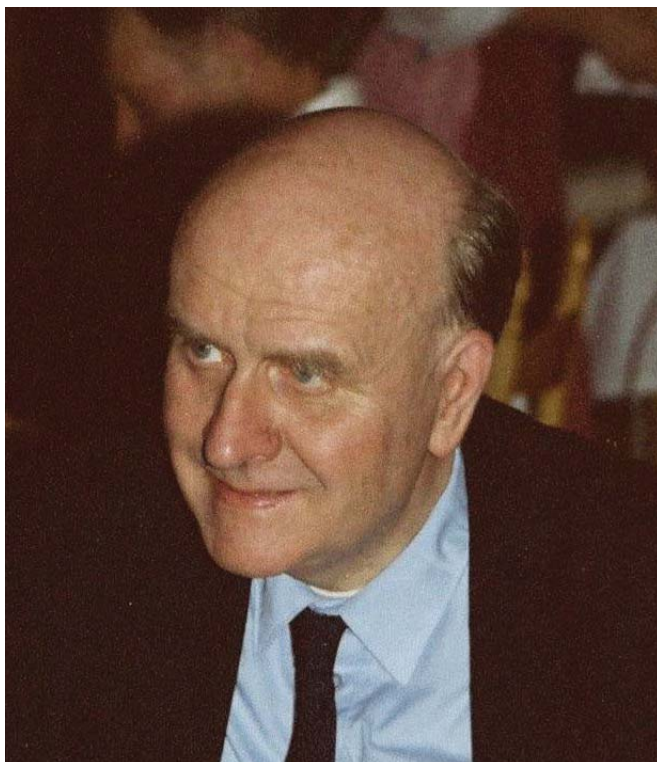
Axel Christian Klixbüll Jørgensen was a “Polyhistor”, one of the very few in the highly specialized science of our time. His interests and contributions in chemistry covered the whole Periodic Table. This statement demonstrates the breadth of his interests, however, it also sheds light on the constraints of chemistry which deals with a large, yet limited number of elements. It is not surprising that Jørgensen went beyond these limits, exploring the probable or plausible chemistry of yet unknown elements and elementary particles such as quarks. Even chemistry itself did not place rigid limits on his mind, he was able to transfer his chemical concepts to scientific problems far beyond the normal such as in astrophysics.

“Structure and Bonding” is intimately associated with the name C.K. Jørgensen both as initiator and author over several decades. The appearance of a special edition in memory of this great scientist is a self-evident prolongation of his many contributions to the success of this series.

We owe a debt of thanks to Dr. Thomas Schönherr for his efforts in setting up this special edition. The scientific contributions reflect some of the outstanding impact on the understanding of chemistry stimulated by Jørgensen’s thoughts as well as by his opening of new research areas. As an innovation in “Structure and Bonding”, a number of personal notes have been added in order to give some impression of the method of thinking and communicating that was so characteristic of Jørgensen. These notes will be a reminder to all those who knew him and they will convey to those who never met him what was so special about this great man.

Stuttgart, September 2003

Arndt Simon



Axel Christian Klixbüll Jørgensen (1931–2001)

Christian Klixbüll Jørgensen

- 1931 Born in Aalborg, Denmark, 18th April
- 1950 Graduation from high school (Abitur). Beginning of studies at the University of Copenhagen. The aim was a *candidatus magisterii* (cand. mag.), which would qualify him as a Danish high school teacher
- 1953 Teaching assistant at the Technical University of Denmark, Copenhagen
- 1954 Cand. mag. in chemistry, mathematics, astronomy and physics with chemistry as the main subject, University of Copenhagen
- 1957 Doctor philosophiae, University of Copenhagen, thesis: *Energy Levels of Complexes and Gaseous Ions, Gjellerup, Copenhagen, Denmark*
- 1957 Marriage to Micheline Prouvez
- 1959 Head of Office for Fundamental Research, Division of the Scientific Businesses, NATO, Paris
- 1961 Director of the Group of Theoretical Inorganic Chemistry, Cyanamid European Research Institute, Cologny, Geneva (until 1968)
- 1962 Books: *Absorption Spectra and Chemical Bonding in Complexes*, Pergamon, Oxford, England. *Orbitals in Atoms and Molecules*, Academic Press, London, England
- 1963 Book: *Inorganic Complexes*, Academic Press, London, England
- 1965 Member of the Royal Danish Academy of Sciences and Letters
- 1966 Editor of *Structure and Bonding* (until 1989)
- 1968 Invited to be Professor at the University of Geneva
- 1969 Book: *Oxidation Numbers and Oxidation States*, Springer-Verlag, Berlin
- 1970 Chair of Physical Chemistry at the University of Geneva
- 1971 Book: *Modern Aspects of Ligand-Field Theory*, Amsterdam, North-Holland
- 1974 Chair of Inorganic and Analytical Chemistry at the University of Geneva
- 1977 Book: *Lasers and Excited States of Rare Earth (with Renata Reisfeld)*, Springer-Verlag, Berlin
- 1978 Death of his wife Micheline
- 1983 Doctor *honoris causa* from the Philosophical Faculty of the University of Zürich
- 1997 Professor Emeritus at the University of Geneva
- 2001 His death on 9th of January

Preface

To honour the late Professor Christian Klixbüll Jørgensen, who was a leading figure in the field of inorganic chemistry, Springer-Verlag agreed to issue a special edition in the review series *Structure and Bonding*. Jørgensen himself initiated the series in 1966 and supported it for almost 25 years as a member of the editorial board. His famous article, “Recent Progress in Ligand-Field Theory” headed the first issue.

Jørgensen carried out small scientific studies even as a schoolboy, and as a student, he often surprised his academic teachers with his enormous knowledge of facts in various fields including astronomy, geography and philosophy and he loved to discuss everything that could be discussed, always with wit and enthusiasm. A characteristic impression of this particular personality is in a letter I received from his friend and early collaborator Claus Erik Schäffer, who recalls the exceptional abilities Klixbüll exhibited more than 50 years ago as a freshman visiting Professor Jannik Bjerrum’s laboratory in Copenhagen at the Technical University of Denmark. This environment embraced only chemists with broad laboratory experience and it had a long tradition in the chemistry of metal complexes. CES: *“In 1950, just after my bachelor’s degree Bjerrum gave me a preparative problem involving a polynuclear chromium(III) ammine complex (saying that he had already had had two students work on it without success). Shortly after I had started working, Klixbüll appeared in the lab and wanted to hear what I was doing. I told him about it, and he immediately responded by telling me that he had “always” (he was a freshman!) found it so curious and almost self-contradictory that chromium(III) amines existed, but yet when precipitating chromium(III) hydroxide with ammonia and adding excess, the precipitate did not redissolve. Why? Then he had tried (in his mother’s kitchen!) to do the experiment when he observed that the filtrate from the hydroxide was slightly red. This led him to do the experiment more slowly and eventually he was able to isolate a red crystalline mass by precipitation with hydrochloric acid. I still have this red high-school sample of his. In that way he took me by surprise. It must have been at about the same time that he read Bjerrum’s thesis overnight and wanted to discuss the contents with Bjerrum. Two years later, when he had finished his bachelor’s degree, he also became a Bjerrum student, and he was assigned the neighbouring cellar room, which he had in fact already used in his spare time to separate the rare earths by fractional crystallisation. On the other side of the corridor, we had a Beckman DU spectrophotometer, which a few years later was replaced by a Cary 11 paid by Marshall Plan money. Under these local geographical conditions, we lived and worked*

side by side - at first as students and later as "assistants", employed by the Technical University - until the end of 1959 when Klixbüll moved to NATO. It was "Sternstunden" (magic moments) for the whole laboratory when all these spectra were unravelled. These "Sternstunden" which included extended discussions of Yamatera's paper in *Naturwissenschaften* (1957) were no doubt fruitful for the forthcoming formulation of the Angular Overlap Model." Today we recognize that this period marked the beginning of a deeper understanding of the electronic spectra of d^n and f^n metal complexes. It was developed on the basis of Jørgensen's encyclopaedic knowledge of inorganic chemistry and atomic spectroscopy, and of his remarkable function as an autodidact in the field of quantum mechanics, even though he remained largely a sceptic with regard to computational efforts in chemistry.

The present volume contains contributions by some of Klixbüll's best friends and collaborators. They describe his human side, along with his profound influence in the fields of optical spectra and chemical bonding in transition metal and rare earth compounds. A biographical note by his longstanding friend Claus Erik Schäffer and an article by Peter Day (The Royal Institution, London) head the volume. Day, who spent post-doctoral time with Jørgensen at the Cyanamide European Research Institute, introduces him as the philosopher and linguist by a fascinating investigation of Jørgensen's contributions to the language of science. A prominent example of this is the "*nephelauxetic effect*" that arises from the decrease in the interelectronic repulsion parameters of the partially filled shell of complexes as compared with the corresponding gaseous ions. This effect belongs to the essentials of ligand-field theory along with the parameterized metal-ligand interactions in terms of the energy parameters e_λ , introduced by Jørgensen and Schäffer as the Angular Overlap Model. This model was a development of the newly designed method, which Jørgensen and his Theoretical Inorganic Group at Cyanamid had created as its publication number 50, to introduce covalency as the explanation for the ligand-field in rare earth complexes. Although such parameters are usually obtained from optical spectra, they can elucidate, sometimes to an astonishing extent (from a rigorous, theoretical point of view), the character of the chemical bonding in the coordination sphere of a transition metal or even of a rare earth compound. In this field Hans-Herbert Schmidtke (Düsseldorf), who together with Pappalardo was involved in this legendary 50th CERITIC paper, contributes to the present Volume with a discussion of the effects that result from the variation of the Racah parameters in highly symmetric complexes. In a complementary manner, Patrick Hoggard, one of the major advocates of the Angular Overlap Model, in his article presents the origin of the AOM parameters and how they may be applied independently of a particular geometry when analysing optical d-d transitions. The way in which the underlying parameters influence the energy level scheme and therefore determine the photon up-conversion processes in Ti^{2+} and Ni^{2+} doped halide lattices is the subject of Oliver Wenger and Hans-Uli Güdel (Bern). An EPR study of Cu(II) complexes is furnished by the group of Harry Gray (Pasadena), who was a postdoctoral fellow with Ballhausen, another former student of Jannik Bjerrum and a significant contributor to the *Sternstunden* in Bjerrum's old laboratory. Harry Gray's co-operative work with Venezuelan colleagues elucidates geometrical and electronic

properties that have been derived from the superhyperfine EPR patterns. Andreas Hauser, who succeeded to Jørgensen's chair at the University of Geneva in 1996, has investigated, together with his group, the influence of chemical pressure on the electronic properties of some tris(bipyridine) complexes that belong to a family of most promising bioinorganic materials. Mihail Atanasov (Sofia) and Claude Daul (Fribourg) have developed a computer-aided ligand-field theory by making use of formalisms and results of Density Functional Theory (DFT). Extensive DFT calculations have been used by Miguel Moreno (Santander) and co-workers to understand pressure effects in cubic and tetragonal copper(II) complexes. A further application of DFT, crossing the border into organometallic chemistry, is presented by Zofia Stasicka (Kraków) and her colleagues who have examined the structure and photochemistry of complexes of iron and ruthenium. The theoretical article by Susanne Mossin and Høgne Weihe, researchers of the younger generation in the best tradition of the Copenhagen school, offers a method to estimate relevant exchange integrals in binuclear μ -oxo-complexes. Jørgensen's expectations about the nature of the chemical bond in an argon compound are reported by Gernot Frenking (Marburg), whose group presents today's answer to yesterday's question by means of a modern theoretical investigation.

Energy levels of rare earth ions and their complexes always had Jørgensen's special attention. In this Volume they are represented by articles from two of his closest friends in his later days. Baldassare DiBartolo (Boston), together with Brian Bowlby, has studied the multiple luminescence of praseodymium-doped BaY_2F_8 , and the last scientific article of this volume is provided by Renata Reisfeld (Jerusalem). She has not only Jørgensen's co-author on a book and a series of papers, but was also Christian's faithful companion at many conferences through the years. Here she presents a paper at the centre of their common interests, an overview of the spectroscopy of rare earth ions in selected luminescent glasses.

The appendix is a special tribute from the community to the man Christian Klixbüll Jørgensen. Herein, the reader will find reminiscences of his particular friends and colleagues. The contributions submitted by Fausto Calderazzo, Baldassare DiBartolo, Brian Judd, Anthony Lucken, George Kauffman, Makoto Morita, Dirk Reinen, Renata Reisfeld, Hans-Herbert Schmidtke and Alan Williams cover biographical details, personal memories, and also some typical anecdotes full of humour. They yield an appreciation of a unique character in the history of chemistry to those who have never had the opportunity to meet him personally.

A few of Jørgensen's legendary hand-written letters (and reviews, likewise!), some photographs, and the complete list of about 400 publications will be available on the Internet as supplementary material. Such documents help to fill in the picture of an unfailingly friendly, extraordinary scientist, who *was aided by a truly phenomenal memory, which enabled him to pluck a reprint from exactly the right pile in his office and carefully correct a misprint in the page numbers of a reference in a paper published twenty years before* (cf. Alan Williams' note). This exceptional ability allowed him to write down the greater part of his scientific work just by putting pen to paper with no further corrections. More important, how-

ever, he was setting down new ideas, founded on an incredible array of facts and fundamentals he had in mind, in combination with an occasionally visionary intuition. He remained faithful to all the disciplines that had attracted his attention in his younger years. In particular, everything that had to do with energy levels of chemical systems, he gave a high rank. Our knowledge about this area today has been influenced more than we know by his contributions. So it remains the particular merit of Christian Klixbüll Jørgensen to have delivered outstanding contributions that have enlarged the foundations and steered the development of modern inorganic chemistry.

As the editor of the Jørgensen volumes in the *Structure and Bonding* series, I am deeply grateful to the authors of the scientific and personal contributions presented here. The response was overwhelming and all agreed with enthusiasm to participate in this project despite the burdens on the shoulders of all of them. Very special thanks are owed to Claus Schäffer for many helpful conversations and for his continuous support. Speaking for myself, I am truly excited to be involved in this edition, because Jørgensen's books and papers as well as the honest and friendly attitude of a surely extraordinary personality have been a grand inspiration throughout my life.

Düsseldorf, August 2003

Thomas Schönherr

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