



Autobiography of Hans-Joachim Freund

I was born on March 4, 1951, about 30 km northeast of Cologne, in Solingen, known as the knife town in the western part of the Federal Republic of Germany. My father, Fritz Freund, who spent 5 years as a prisoner of war in Russia after he was captured, barely having survived the battle of Stalingrad, only returned to Germany around Christmas 1949. He and my mother, Lieselotte Freund, née Willms, got married in April 1950, and I started to glimpse at the world almost a year after and remained as the only child. I should have been several years older since my parents had known each other since their elementary school years, just after World War I was over, and they already had plans to get married when the second war started. I lived together with them and both grandparents in a home my grandfather on my mother's side had built in 1911 based on money he earned through his skills making razor blades, typical Solingen craftsmanship. My parents worked within the little company my grandfather had founded, based on manufacturing razors, and they developed it into producing manicure and pedicure tools. My father was originally trained as a blacksmith and propelled himself to attend an engineering school and even university, without finishing because the war broke out. My mother attended the academy of arts in Düsseldorf after high school and became a sculptor. I grew up in a very liberal, arts-friendly atmosphere, but the message was conveyed to me very clearly: If you want to get anywhere, make sure you work for it! Even though the family income was solid, my parents—and I agree with them—always cherished the fact that education was, and still is, free in Germany.

When I was 6 years old, I entered elementary school and after 4 years had to take an entrance exam to enter into Gymnasium. To my parents' delight, I passed. Early on, I developed an interest in biology and chemistry, mainly because I had a fantastic biology teacher, Franz Xaver Mattauch (he was bohemian and had an interesting accent), telling us about Watson and Crick, who received the Nobel Prize in 1962, and about DNA. I was fascinated by the idea that one may understand complex phenomena on the basis of atoms bound to build up molecules. I started to set up a chemistry laboratory at our house, and, if I remember correctly, my parents were not always happy with the chemicals I used. I went to work during the summer breaks to get some money so I could buy some lab gear. The most expensive tool was a Kipp's apparatus to produce gases. When I was not working in my laboratory, I was playing tennis, participating in tournaments and having a lot of fun. More and more, I got interested in understanding the physical principles behind the formation of molecules. This got me interested in quantum mechanics, and my physics teacher pointed me toward some interesting literature that I could understand with the mathematics we had been exposed to by 1967.

In 1969, I passed the final examination, which opened the path to university. It was clear that I would study physics and chemistry, and I inscribed in both courses. After my first semester, in April 1970 at the University of Cologne, I was

drafted to serve in the air force for 18 months. Fortunately, after the first 3 months of basic training at Pinneberg near Hamburg, I was transferred to a unit near Cologne, directly associated with the Ministry of Defense, led by the future chancellor of the Federal Republic, Helmut Schmidt. It was the Search and Rescue Unit (SAR), where we worked in shifts. This allowed me, by personal permission from the minister, to keep my studies up during my military service. In 1972, I received the B.Sc.-equivalent "Vordiplom" in both fields and continued during the Master's studies to be interested in both, but in late 1974, I wanted to perform my experimental Master's work (Diplomarbeit) in physics with Herbert Walther, who, however, moved from the Physics Department in Cologne to the University of Munich and later set up the Max Planck Institute for Quantum Optics. At that time, I had just met my future wife, Susanne Herfurth, and I decided to stay in Cologne. I finally opted to do my experimental Master's work with a physicist, Georg Hohlneicher, who came from the Physical Chemistry Department at the Technical University of Munich and had taken a position in the Chemistry Department, i.e., in Organic Chemistry. He gave lectures on photoelectron spectroscopy as an experimental tool as well as numerical quantum chemical calculations for electronic properties, and I got interested. Georg Hohlneicher purchased a spectrometer from Leybold-Heraeus, the famous and brand new LHS-10. Together with my colleague Horst Gonska, who also had just started to work in Hohlneicher's group as a staff member, we worked our way through the "teething troubles" of the machine and its electron optics, and by mid-1975 I had finished my diploma work on charging of insulators in X-ray photoelectron spectroscopy. It took a few months to sort out this unusual physics/chemistry situation and which department would actually accept the diploma thesis, but, finally, by the end of 1975, I was set and could think about starting the work for my Ph.D.

In late summer 1975, I participated in the Quantum Chemistry Summer School organized by Per-Olof Löwdin in Uppsala, Sweden. The participation was supported by a prestigious organization called "Studienstiftung des Deutschen Volkes", awarding me a fellowship after my B.Sc. examination, which allowed me to rent a small apartment close to the Chemistry Department in Cologne. Before, I had taken the train every day back and forth between Cologne and my hometown Solingen. The summer school was an eye-opener for me as I realized the power of quantum chemical calculations. It also served as a platform to meet young colleagues, who later turned out to become prominent experimentalists and theorists. There was another eye-opening experience at the time: In order to explore how it would be to become a high school teacher, between 1975 and 1978, I worked as an "Aushilfslehrer" in my old Gymnasium in Solingen. That was possible because at the time, teachers for

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natural sciences were in high demand, and there was simply not a sufficient supply of teachers. Therefore, when a student in physics or chemistry received his Diploma, he was simultaneously given the "Erstes Staatsexamen", which allowed him to teach at a high school toward the second exam, the final approval to work as a teacher. This work made me really appreciate how much hard work it is to be a teacher and how important it is to get young people seriously interested in a topic.

While thinking about my Ph.D. and digesting all the impressions from Sweden, I wrote drafts of the first two papers on the topic of my diploma work. Those were finished in spring 1976, then thoroughly commented and corrected by my advisor, sent by surface mail to the editorial office of the *Journal of Electron Spectroscopy and Related Phenomena*, and printed in 1977 in the above journal. By that time, the topic of my thesis work started to take shape: There were ideas around suggesting that molecular binding to metal surfaces should be looked at as an electronically local event similar to the situation found in metal complexes. Analogies from infrared spectroscopy had been used to support the idea, and so it was near at hand to see whether one might find similar correspondences in photoelectron spectroscopy. Those ideas were obviously widespread: Gerhard Ertl's group in Munich had similar ideas looking at CO on a Pd(111) surface and published a paper in *Chemical Physics Letters* in 1976 on this. I was interested in expanding this idea to other surfaces and decided to work on Co(0001) to systematize the effort. I bought the crystal with my own funds received through my fellowship, aimed at buying books. The first thing I did was sputter and heat it, only to find out that the thermocouple was not well-connected and I had heated it above 400 °C, which leads to a phase transition from hcp to fcc, and the single crystal was gone. My thesis advisor, after some discussion, provided a new one. We took photoelectron spectra of CO adsorbed on the surface and compared them with the corresponding spectra of a series of cobalt carbonyls, i.e., $\text{Co}_2(\text{CO})_6$, $\text{Co}_4(\text{CO})_{12}$, and $\text{Co}_6(\text{CO})_{16}$. This series had the advantage that we could vary the metal content. The C 1s spectra showed what turned out to be interesting spectral functions exhibiting pronounced shakeup satellites. Similar phenomena were also found in the range of the inner valence ionizations. The experimental data needed to be interpreted and were, eventually, but only after my habilitation in 1983 (see below). The reason was that in order to understand the photoelectron spectra, we had to develop an electronic structure code to calculate them. Handling transition-metal complexes of the size we had in mind using ab initio methods was not possible at the time. We resorted to a semiempirical code called CNDO (which stands for Complete Neglect of Differential Overlap), and I modified it to include transition-metal elements. Then, we basically used Koopmans' Theorem to interpret the spectra in the valence regime. We concluded from a comparison between experimental spectra of the complexes mentioned above and theory that some of the trends in electronic structure, suggested by the calculations and observed experimentally, when varying the metal content could be correlated. Encouraged by that, I expressed the hope that by a deeper understanding of the spectra we would be able to get insight into factors controlling the interaction of molecules on surfaces and thus contribute to understanding reactions at surfaces and in heterogeneous catalysis. Of course, a deeper understanding of the satellite features, alluded to above, could not be expected to be

described on the basis of an independent single particle picture. A comment on the side: Another student in the group, Bernhard Dick, who is now a Professor of Physical Chemistry at Regensburg University, implemented a singles and doubles configuration interaction code where the wave functions generated by my code could be used to calculate electronic excitation spectra.

When I was halfway through my thesis, in summer 1977, I got married to Susanne Herfurth. On our honeymoon, we took our VW Beetle from Cologne across the channel to the U.K., specifically to Wales, and after touring Wales, we went back to the continent, rested along the Loire River in France, and finally visited my parents-in-law in the Alps in Switzerland, where they used to go for vacation. Shortly after we returned to Cologne, I left by train for my first two conference attendances, i.e., the 5th Vacuum Ultraviolet Radiation Physics Conference in Montpellier, France and after that the 7th International Vacuum Congress in Vienna, Austria. Those were fantastic experiences that left a lasting influence. At the end of 1978, I defended my thesis "summa cum laude", and I discussed with my thesis advisor, Georg Hohlneicher, as well as with the Professors in Physical Chemistry in the Cologne Chemistry Department whether they would support my attempt to go for a Habilitation in Physical Chemistry within the 5 years to come. The response was very positive, and I was assigned the formal position of "Wissenschaftlicher Assistent". So, I immediately, started to try to get deeper insight into the problem of how to describe the above-mentioned satellite structure in photoelectron spectra of adsorbed molecules. One of the Ph.D. students in the group, Dietmar Saddei, who is now a Senior Vice President of SAP and who I had to mentor as a habilitant, was asked to create a code to perform second-order Tamm–Dancoff calculations on the ground-state wave function of the complexes, which would allow the direct description of the photoelectron spectra in the valence and inner valence regime. The methodological concept of the 2nd Tamm–Dancoff approximation to be used to calculate photoelectron spectra had been developed by Lorenz Cederbaum when he was in Georg Hohlneicher's group in Munich. This worked out very well and led to a number of publications.

Around this time, my mentor had heard about studies on shakeup structures of complexes in the U.S. by Ward Plummer at Penn and Bill Salaneck at Xerox Research Laboratories in Webster in upstate New York. I contacted them, and Ward agreed to take me on as a postdoc. I received a fellowship from the German Science Foundation (DFG), and Susanne and I left for Philadelphia in late 1979. Susanne gave up her job when we decided to go. At the time, she was heading a childcare near the place we lived, in a small apartment in Cologne. The physics department at Penn was a great place with wonderful lunch break meetings. To live in West Philly at the time, however, was not easy to adapt to as a German who had not been exposed to crime scenes. I bought a used Plymouth Horizon car, for which I had to pay exorbitant insurance fees living there. Working with Ward was very rewarding scientifically, and we also got along very well on the personal side. Susanne and I eventually moved to Webster to work at the Xerox Laboratories. We lived in Rochester and were very lucky with our apartment on East Avenue, right next to the Eastman House. The fees for the car insurance dropped by a factor of 4. Working with Bill Salaneck, Charlie Duke, and Dick Bigelow was great fun. Ward visited from time to time,

and I participated in beam times at the Synchrotron in Stoughton, close to Madison, Wisconsin. At “Tantalus”—the name of the synchrotron—I met many colleagues who I had contact with during my later scientific life: Wolfgang Eberhard, Felix Greuter, Torgny Gustafsson, Peter Heinemann, Franz Himpsel, and many others. I cherish my times at Penn and Xerox. They were key for my future career. A number of publications on adsorbed CO on metal surfaces, including adsorbate band structures, and on the interpretation of the satellite structure resulted from the work with Ward and my colleagues, which I planned to use for my habilitation project back in Germany. Susanne and I had a great time in the U.S. We took a camper trip from the East Coast to the West Coast through the South of the U.S. and back through the North. It was a wonderful experience we never forgot. Before we moved back to Germany in 1981, we had a trip to Florida and all over the Keys. After we returned, I continued to work on my habilitation in Cologne, and 9 months after our Florida trip, Julia, our first child was born. I am very happy that I have kept contact with many I met during my time there, with Ward Plummer, in particular!

In the summer of 1982, I spent 2 months at Brookhaven with Wolfgang Eberhard, who had moved there from Penn. In June 1983, I submitted my Habilitation thesis in Physical Chemistry, not realizing that by then the relation between my mentor and the leaders in Physical Chemistry had deteriorated. At the same time, I was looking for a job to feed my family. Among several other applications, I also applied for an Associate Professorship at the University of Erlangen-Nürnberg. When the problems between my mentor and the colleagues culminated and I was worried that my habilitation would not be accepted, I received a phone call from Gerd Wedler, the Professor of Physical Chemistry with whom my position would be associated, who declared that the University of Erlangen-Nürnberg would offer me the job even without the formal habilitation. A few days later, the famous theoretical physicist Bernhard Mühlischlegel in the Physics Department in Cologne called my mentor and offered to award my habilitation in physics so that the problems in the Chemistry Department would become irrelevant. A week later, I passed the lecture examination in front of the entire faculty and was awarded the title Dr. rer. nat. habil. and Private Docent in Chemical Physics, not in Physical Chemistry. In June 1983, I was offered the job in Erlangen, and in November of the same year, I started teaching and setting up my research group there. Susanne and I rented a house in the small village of Dormitz close to Erlangen, and Julia, now 2 years old, went to childcare in Franconia—the part of Bavaria we lived in—and developed a clear language accent. The job in Erlangen was an enormous opportunity for me. I developed new research lines on CO₂ adsorption and activation. In this connection, another person I had met during my time at Penn became important for me: Dick Messmer, a theorist working at General Electric. I spent the summers of 1984 and 1985 as a fellow at GE in Schenectady, New York working with Dick, specifically on CO₂ activation. When in Erlangen, I developed strong relations with the solid-state physics groups led by Klaus Müller and Klaus Heinz. My first Ph.D. student in Erlangen came from physics: Bernd Bartos. Klaus Müller and Gerd Wedler initiated what is called a “Forschergruppe”—a small collaborative research center—called “Reactivity at Surfaces”, which received funding from the German Science Foundation in 1985, and I was part of it and was able to establish my own

research lab and set up the first ultrahigh vacuum instrument of my own. We started research projects using synchrotron radiation at BESSY in Berlin and initiated collaborations with other groups in Germany interested in surface studies. At that time, surface physics and chemistry were popular fields here, and there was a very active community. Ward Plummer, who I nominated together with Gerd Wedler for a Humboldt Award, spent a few months in Erlangen with us, a time I very fondly remember.

The time in Erlangen was extremely rewarding for me, and by late 1986, I applied for a chair at Ruhr-University of Bochum. I got short-listed, and I was offered the job in 1987. I moved to Bochum in the summer of that year. The year before, in November 1986, my twin sons, Martin and Sebastian, were born in Erlangen. We bought a house in Bochum, and I started to build up a group at the University. I was extremely lucky: I inherited an excellent group of technical and scientific staff from my predecessor, Heinrich Richtering, on the Chair of Physical Chemistry at the Ruhr-University Bochum, among them Heiko Hamann. He helped enormously to recruit many excellent students to join the group by organizing a tea meeting to explain to them our research strategy. Unusual for chemistry curricula at German universities, in Bochum, students had to take physical chemistry starting with the first semester. He identified the good students during those courses and got them interested in joining us. Marcus Bäumer and Jörg Libuda were among them. We started research projects involving synchrotron studies. I also had the fortune to meet Manfred Neumann from Osnabrück, a former student of Gerhard Ertl, who had a student, Helmut Kühlenbeck. We interacted closely on his thesis project, and after his Ph.D., Helmut Kühlenbeck joined my group in Bochum as a staff member. Based on the experience I had from my time in Erlangen, we developed a concept for a “Forschergruppe” in Bochum. Helmut had started to work on preparing oxide surfaces by growing epitaxial oxide films of nickel oxides on single-crystal Ni substrates. At that time, very few groups were working on oxides. Vic Henrich at Yale and Claudine Noguera in theory were two of several. The idea was to use the oxide substrates as reactive substrates and as supports for metal nanoparticles; we would prepare them via chemical vapor deposition and create model systems for heterogeneous catalysts. We continued to work on CO₂ activation, now on oxides, in contact with the Wyn Roberts group in Cardiff. We proposed to the German Science Foundation to fund a “Forschergruppe” on model catalysts. The evaluators, among them Gerhard Ertl and Ernst Bauer, thought that was a good idea, and we got funded in 1989. Manfred Baerns and Helmut Papp from Technical Chemistry and Henning Neddermeyer from Physics in Bochum were part of it, as well as Volker Staemmler from the Theoretical Chemistry group. I had the fortune to spend a sabbatical at Berkeley with Gabor Somorjai in 1990, and on that occasion I met Wayne Goodman, who apparently was working on very similar ideas. We became and remained very good friends until he sadly passed away in 2012. Wayne was the second Humboldt Awardee that I nominated, and he came over to visit us in Bochum and later in Berlin. The funding of the “Forschergruppe” carried us until 1994, and studies in model catalysis in its spirit became popular. In 1992, I was offered a chair in physical chemistry at the University of Freiburg, and I was tempted to leave Bochum, but the funding situation was so much better here that I finally decided to stay.

To my big surprise, in 1995, the German Science Foundation awarded me the Gottfried Wilhelm Leibniz Prize, considered to be one of the prime science awards in Germany. In the same year, I was offered my current position at the Fritz Haber Institute as a successor of Jochen Block. I was very happy that almost all my students and postdocs, including the two permanent scientific staff, Heiko Hamann and Helmut Kuhlenbeck, agreed to join me. We started to set up the new Department of Chemical Physics. A new building was planned and erected in record time due to the fact that Heiko Hamann took care of all organizational aspects so the other members of the group could concentrate on science. My colleagues, Alex Bradshaw, Gerhard Ertl, Matthias Scheffler, and Robert Schlögl, were extremely helpful in the entire process, and we got going fast. In the following 22 years, I enjoyed working at the "Fritz", also with the new colleagues, Gerard Meijer and Martin Wolf. Twice I had been offered jobs at other places, but I decided to stay.

In 2004, my wife Susanne, who had been the backbone of the family, tragically passed away after suffering from a brain tumor. This, of course, was a difficult and critical time to keep the children on track and the family stable. I thank my daughter, Julia Freund (now Julia Eberl after marrying Sebastian Eberl, having two children, my grandchildren, Paul (3) and Johanna (1)), my sons Martin and Sebastian for being strong and stable, and all my friends for supporting me. Fortunately, a new person came into my life: Tatiana Dutkova. We got married in 2007, and she brought a fourth child into the family: Jan. I am very grateful for her support and love over the years.

In the same year, I became a founding member of the European Research Council in Brussels, which set up a completely new research funding style for the European Union based only on the track record of the person, the quality and originality of the proposed project, and trust. This has been a rewarding experience. The work in the Department covered a broad range of topics mostly connected with oxides, model catalysts, and reactions, both thermal as well as photoinduced, but also including topics within general physical chemistry, such as the transformation of crystalline and vitreous oxides, specifically silica. We also invested in the development of experimental techniques, such as electron spin resonance spectroscopy at surfaces and aberration-corrected spectromicroscopy (SMART LEEM-PEEM) using synchrotron radiation. Some of this was in collaboration with other colleagues, in particular, the latter with Eberhard Umbach. The collaboration with the Berlin Universities (FU, HU, TU) has been fantastic. I cannot think of a better and more fruitful environment between a Max Planck Institute and the surrounding universities. I have, at the same time, profited from many contacts to leading scientists around the world and collaborations with them, for example, Kiyotaka Asakura, Micha Asscher, Ernst Bauer, Charlie Campbell, Ulrike Diebold, Cynthia Friend, Hongjun Gao, Wayne Goodman, John Hemminger, Claude Henry, Weixin Huang, Maki Kawai, Bob Madix, Ted Oyama, Martin Schmal, Fernando Stavale, Peter Stair, Hans-Peter Steinrück, Christof Wöll, and, last but not least, Gabor Somorjai, just to name a few, who have provided inspiration and good advice and have been lifelong friends. I spent a couple of sabbaticals with my old friend John Hemminger at the Chemistry Department at Irvine, which also allowed me to visit Wilson Ho, who I met first when I came to Penn and he had just left for Cornell after his Ph.D.

with Ward in the Physics Department. One other sabbatical I spent at Berkeley with Gabor and another one with Claude Henry at Luminy in southern France near Marseille.

There have been many more important contacts in my scientific life that I should mention. There is one aspect, in addition, I want to explicitly underline: our joint efforts and fruitful collaborations with theory groups. Much of our work would have considerably less impact or would have been impossible had it not been for interactions with Paul Bagus, Gianfranco Pacchioni, Joachim Sauer, Hannu Häkkinen, and Notker Rösch. I am very grateful for their personal friendship and to them and their collaborators for the truly open and trustful joint efforts, which I truly cherish! Many of the scientists mentioned have provided their support of my scientific endeavors through nominations and letters of support for the awards and distinctions I have been so fortunate to receive. Thank you very much! I would like to dedicate a special thanks also to those scientists who led working groups within the Department of Chemical Physics (partially accompanying me from Bochum) over the years: Katharina Al-Shamery, Marcus Bäumer, Wolfgang Drachsel, Norbert Ernst, Heiko Hamann, Markus Heyde, Heinz Junkes (PP&B), Thorsten Klüner, Helmut Kuhlenbeck, Jörg Libuda, Niklas Nilus, Thomas Risse, Günther Rupprechter, Hans-Peter Rust, Svetlana Schaueremann, Thomas Schmidt, Shamil Shaikhutdinov, Martin Sterrer, and Kazuo Watanabe. There is an institution I would like to thank as well: the Alexander von Humboldt Foundation! It has funded and supported a large number of junior and senior scientists in their association with the Chemical Physics Department. Some of them have been addressed above. The list of fellows and awardees is too long to be included here. I thank each and every one of them for their support. A special thanks goes to the so-called Postprofs (a term invented by Karl Heinz Rieder for retired Professors), who contributed so much to the success of the Department and have been personal friends and advisors: Dietrich Menzel, Karl-Heinz Rieder, Wolf-Dieter Schneider, and Eberhard Umbach. Without the excellent technical staff at the institute, most of the work would have been impossible. The same applies to my long-time secretaries, in particular, Karin Klug and Manuela Misch, for the enormous help I received during my professional life. At the end, I would like to thank the most important set of people in my scientific life: all the students and postdocs. Without them, nothing would have been achieved. I am extremely grateful to all of them and the entire worldwide surface science and catalysis family for accepting me as their member. Thank you!!!!

Hans-Joachim Freund