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Special Issue Preface

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Autobiography of Wolfgang Lubitz: A Professional History[†]

EDUCATION (1969−1974), GRADUATE STUDIES AND HABILITATION (1974−1979) AT THE FU BERLIN

I was born in West Berlin in 1949, where I also went to school. During high school, I enjoyed studying, in particular the sciences, where I had very good teachers. Early in 1969, I graduated and with the summer semester started to study chemistry and physics at the Free University (FU) Berlin. I obtained my Diploma in 1974. My diploma thesis I did in physical organic chemistry with Professor Harry Kurreck, a fantastic teacher and scientist, who worked on the synthesis and spectroscopy of organic radicals, mainly using EPR techniques. He offered me to join a collaborative project with his friend Professor Klaus Möbius from the physics department of FU Berlin. I started to work on fluorinated aromatic radicals that were difficult to prepare and needed a chemist. The idea was to apply the rather new electron−nuclear double resonance (ENDOR) technique to such radicals in liquid solution and detect not only the proton but also the fluorine isotropic hyperfine couplings, necessary to obtain spin density distribution maps of these molecules. The compounds came mostly from Herbert Zimmermann, an exceptionally gifted chemist who worked as a technician at the Max Planck Institute in Heidelberg with Karl Hausser. With Herbert, I continued to work for many years and we developed a friendship that is lasting until today. The ENDOR spectrometer we used was entirely home-built by Klaus Möbius and his students Klaus-Peter Dinse and Reinhard Biehl. It was difficult to use and suffered from stray-field problems due to the simultaneously irradiated very high microwave (mw) and radiofrequency (rf) power levels in the resonator that were necessary to perform these experiments. Often we had to work at night when the performance of the instrument was much better. During this time, my direct supervisor was Klaus-Peter Dinse. With Peter and his family, we later started to have frequent contacts and met on many scientific meetings for several years.

The detection of ^{19}F ENDOR was successful, and I proudly published my first paper on this topic.[1] I found myself in a comfortable situation as interface and contact person between the chemistry and physics departments and was offered to stay for my doctoral work. I became a teaching assistant in chemistry, earned good money, and rented an apartment close to university. The following years were probably the most relaxed ones of my life. I liked teaching students in chemistry and worked mostly in physics for my thesis—but also helped to set up the first ENDOR instrument in the Chemistry Department of FU Berlin.

During my doctoral thesis, I developed high-power solution ENDOR for nonproton nuclei. Many of the isotopically labeled compounds came from the lab of Harry Kurreck, with whom I had a very nice personal relationship. In the 1980s, we wrote a book together with Burkhard Kirste on *ENDOR Spectroscopy of Radicals in Solution*,[44] which became a standard work in the field. Early on, we recognized that ENDOR of proton and especially nonproton nuclei required a solid theoretical framework. Martin Plato, a staff scientist of the group of Klaus Möbius, was interested in this problem. On the basis of the early work of Jack Freed (Cornell University), he developed a computer program by which one could calculate ENDOR spectra of a given radical (line intensities, line widths, saturation behavior, coherence effects, etc.) as a function of several parameters (radical concentration, solvent viscosity and temperature, field strengths, and other instrumental parameters). This enabled us to estimate the optimal experimental conditions for detection of a particular nucleus in any given radical. This work was published in *J. Phys. Chem.* in 1981.[13] It gave me enormous pleasure to work with Martin on this project, and I have profited a lot from it. We later continued our collaboration doing quantum chemical calculations on different radicals, e.g., on complicated chlorophyll radicals. To recognize the important interplay of spectroscopy and quantum chemistry was crucial for my later career, where we sought to underpin our results in many cases by calculations, either with theoretical chemists of my own group (Matthias Stein, Sebastian Sinnecker) or through collaborations, in particular with Frank Neese (Mülheim). It is very clear that many complex spectroscopic problems cannot be solved and fully understood without theoretical support.

My experimental work would not have been possible without the ingenious concept developed by Reinhard Biehl for constructing a new type of ENDOR resonator.[2] This provided higher stability and sensitivity, which, for example, even allowed the detection of ENDOR resonances of rare nuclei like ^{13}C (1.1%)[8] and ^{29}Si (4.7%)[26] in natural abundance. I was fortunate to be present when these developments in the laboratory of Klaus Möbius took place. It was a very productive and stimulating time, and I learned a lot from the daily work with Reinhard, who at this time also developed and implemented electron−nuclear−nuclear triple resonance techniques. Later, he moved to Bruker, where he built the first cw-EPR/ENDOR/TRIPLE spectrometer, which laid the foundation for the success of Bruker-EPR/BioSpin headed by Dieter Schmalbein. It was very sad for his family and friends that Reinhard died so early (in 1987) at the age of only 43.

I finished my doctoral work in 1977, which was sufficiently interesting that my supervisors nominated me for the prestigious "Otto-Klung-Preis" of the FU Berlin, which I received in 1978. In December 1977, I visited the USA (New York City) with my girlfriend and later wife Gisela for the first time and came back full of impressions and excitement. I was planning for a postdoctoral position in the USA, but Klaus Möbius and Harry Kurreck convinced me to stay in Berlin and go to the USA at a later more advanced stage of my career. In

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1979, I received a position as Assistant Professor at the Chemistry Department of FU Berlin and started to teach organic chemistry, quantum chemistry, and spectroscopy.

■ ASSISTANT PROFESSOR AT FU BERLIN (1979−1986)

With the knowledge and experience of my doctoral work, I thought about new applications of the EPR and ENDOR techniques. In 1977, Arnold Hoff from Leiden University visited the Möbius laboratory to perform experiments on photosynthetic pigments. I had long discussions with Arnold, and he convinced me that "Photosynthesis is a Garden of Eden for an EPR Spectroscopist", as he expressed it in one of his review articles. Together with Friedhelm Lendzian, a doctoral student in the lab of Klaus Möbius, I started to study chlorophyll radical ions occurring in the electron transfer (ET) chain of bacterial and plant photosynthetic reaction centers (RCs) using ENDOR-in-solution. We were able to resolve and assign the vast majority of the nuclear hyperfine coupling constants, thereby expanding and adding to earlier work in this field, e.g., by Jack Fajer (Brookhaven, USA). Shortly after this time, we established cooperation with Hugo Scheer, an expert in photosynthesis at the University of Munich. He supplied us with RCs of photosynthetic bacteria of very good quality and helped with sample preparation. My collaboration with Hugo continued over several decades, and we still enjoy meeting with our families. On the RC samples, we successfully performed high-resolution cw-ENDOR under physiological conditions on the light-induced radical cation of the primary donor in this large membrane protein. By comparison with the bacteriochlorophyll (BChl) radical cation in solution, we showed that the primary donor is a BChl dimer in which the spin/charge density distribution is asymmetrically distributed over the dimer halves,[15,27] which was later corroborated by single-crystal ENDOR studies[72] and quantum chemical calculations performed together with Martin Plato. At this time, the semiempirical approach developed by Martin (RHF-INDO/SP) was state-of-the-art for the calculation of large open-shell molecules. The work was later summarized in two articles in Hugo Scheer's CRC book *Chlorophylls*.[58,59] The described work established the position of our laboratory in the photosynthetic community. Around this time, the first crystallization and structural analysis of a bacterial reaction center was reported that was a few years later (1988) honored with the Nobel Prize in Chemistry for Hartmut Michel, Johann Deisenhofer, and Robert Huber (MPI for Biochemistry, Martinsried). This was an exciting time for all of us working on bacterial RCs. Mainly based on the work on photosynthetic pigments and reaction centers, I received my "habilitation" in 1982 and became a lecturer ("Privatdozent") at the FU Berlin.

Already a few years earlier (1979), I got married to my long-time girlfriend Gisela. To our great pleasure, in 1981, our son Felix was born with whom we had so much fun and still have a wonderful relationship. Together we decided to go to California. During previous trips in 1979 and 1982, I had visited the laboratory of George Feher at the University of California San Diego in La Jolla, and after consultation with Arnold Hoff and Klaus Möbius, George accepted me in his laboratory.

■ RESEARCH SCIENTIST AT UC SAN DIEGO (1983−1984)

My stay at UCSD (June 1983 to December 1984) was supported by a stipend from the Max Kade Foundation (New York). I was actually George's first postdoc from Germany, and it took many years before he started to tell me his personal story and about the lasting conflict he has with Germany and the Holocaust. My discussion with George is now continuing for more than 30 years, and the outcome deeply impressed me and my family. Today I can say that this experience became an essential part of our lives. After our stay in USA, I visited Israel together with the family. We saw Yad Vashem in Jerusalem seeking to understand the unbelievable that has happened in our country some 40−50 years ago. We made friends in Israel, and I started several joint projects in the following years with groups in Jerusalem (Itamar Willner, Rachel Nechushtai), Haifa/Technion (Noam Adir), and the Weizmann Institute in Rehovot (Avigdor Scherz, Daniella Goldfarb, Lev Weiner); for the Minerva Foundation and other organizations (GIF, DIP), I have been many times reviewer or project partner during the last 20 years.

During the time at UCSD, I continued to work on bacterial RCs and related pigments. Together with Roger Isaacson, a very gifted microwave and radiofrequency engineer and excellent lab manager and scientist, we built different variants of EPR/ENDOR resonators and soon started also solution cw-ENDOR experiments in La Jolla. Over the years, I developed a lasting friendship with Roger and his family. They visited Germany several times, and our close relationship is still continuing. We did many exciting scientific experiments together. For our laboratory in Germany, the profound knowledge of Roger in instrument design (both X- and Q-band) and resonator development was very helpful.[334] During the time in California, I also learned how to grow bacteria and prepare and crystallize photosynthetic RCs. We specifically isotope labeled the RC and the pigments and prepared various radical cations and anions. This work was done with George's technician Ed Abresch to whom I owe most of my skills in biopreparations. At this time, James Allen and his later wife JoAnn Williams were also in George's lab, working on crystallization of RCs and doing the first RC protein sequencing and later also site-directed mutagenesis of RCs. They both went to Arizona State University in Tempe to continue their excellent work, and we enjoyed a very fruitful collaboration with them over many years in the 1990s.

Triggered by discussions with George Feher and Mel Okamura, I started already in 1983/84 to also work on the quinone acceptors in the RC. Mel showed me how to prepare the radical anions of Q_A and Q_B separately with laser flashes, and I learned how to remove and reconstitute quinones, how to exchange the coupled Fe^{2+} (e.g., with Zn^{2+}), and how to label and substitute the quinones. Later we worked on Q_A and Q_B also in single crystals, and we got information about the important H-bonding by means of ENDOR. With Mel, I had a special relationship. We have been traveling together to interesting places in the USA, Japan, England, France, and Germany, and later in his career, Mel spent a sabbatical with us in Mülheim, Germany. With his wife, daughters, and family, we have many ties that last until today.

The major results on the quinones were summarized in a review[147] written during the sabbatical I spent with the family at UCSD 1997/98. However, finally finished was this difficult

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project by a postdoc from Peru, Marco Flores, over 20 years after the initial experiments.[236,247] Marco first worked with George at UCSD and then several years with me in Germany. The time in La Jolla had a profound influence on my further work, and I am very grateful to the Feher group for sharing ideas, for helping with difficult preparations and instrument problems, and for a continuing friendship with many group members over more than three decades.

■ ASSOCIATE PROFESSOR AT FU BERLIN (1986−1989)

The experience gained both at UCSD and previously in the physics laboratory of Klaus Möbius and in the synthetic chemistry group of Harry Kurreck enabled me to build my own group after returning to FU Berlin. As a young professor in organic chemistry, I enjoyed support by the institute and soon joined a collaborative research center (Sfb 312) of the DFG ("Vectorial Membrane Processes") chaired by Jürgen Fuhrhop at the FU Berlin, with a project on bacterial photosynthetic RCs, using mostly EPR and ENDOR techniques. Influenced by the excellent work of the groups of Gernot Renger and Horst Witt from the Max Volmer Institute at the Technical University (TU) Berlin on oxygenic photosynthesis, we started to collaborate with them within the Sfb on Photosystem (PS) I and PS II. In 1988, I got two offers for permanent positons as Associate Professor at the University of Osnabrück and Stuttgart. I accepted the offer from Stuttgart.

■ ASSOCIATE PROFESSOR AT UNIVERSITY OF STUTTGART (1989−1991)

The position was in Experimental Physics. The Department was looking for somebody to teach biophysics and establish a related group, and the choice was on me. I started teaching a biophysics course early in 1989. My family was still in Berlin for half a year and then followed me to Vaihingen/Stuttgart. Our son Felix spent two years in primary school in Vaihingen; he took the change quite well and learned a lot in the different school system in the state of Baden-Württemberg. During this time, one of the most profound changes happened in Germany and Europe—the Berlin Wall was opened on November 9, 1989. This was a consequence of the economic and political breakdown of East Germany, and it also marked the beginning of a major change in East Europe and the Soviet Union.

Shortly before this event, the Berlin magnetic resonance groups of Klaus Möbius, Dietmar Stehlik, Harry Kurreck, and myself with several of our students visited Moscow, Novosibirsk, Kazan, and Leningrad on an extended journey to strengthen old and initiate new collaborations with the country, where magnetic resonance was invented as early as 1944 by E. Zavoisky (Kazan). In the Soviet Union, much good EPR developments and applications had been done of which many of us had not the faintest idea, since most of the work was not published or appeared only in Russian journals. This trip marked the beginning of a long intensive exchange and collaboration with Russian scientists that was very important for our instrumental and methodological development in the forthcoming years and still is today. Later I returned many times to Russia—in particular to Kazan, where a good friend Kev Salikov is the director of the Zavoisky Physical-Technical Institute of the Russian Academy of Sciences. He also founded the first English journal *Applied Magnetic Resonance* in Russia, and in his institute, the *EPR Newsletter* of the International EPR/ESR Society is edited by Laila Mosina in a truly professional and excellent way. I enjoyed a close collaboration with her especially during my years as President of the Society (2005−2008). One of the nicest events in Kazan for me and my wife was the wonderful celebration of the Zavoisky Award that I received in 2002.

In Stuttgart, I was a member of the second Physical Institute of the university headed by Michael Mehring. He—as an NMR expert and very interested in related techniques—was actually one of the pioneers of pulsed EPR, maybe best demonstrated by the invention of HYSCORE spectroscopy, published together with his student Peter Höfer in 1986. Peter went to Bruker in Karlsruhe as the successor of Reinhard Biehl, and he is now the head of the EPR section. My group owes him many good advices and help with instrumental problems and developments. In Stuttgart, I had three very talented doctoral students (Hanno Käss, Wolfgang Zweygart, and Joachim Rautter) who built up the laboratory and did first experiments. We profited a lot from the excellent Mehring laboratories, the knowledge of the dedicated workshops, and the kind support of Michael in any respect. However, my stay in Stuttgart was not lasting long; already in 1991, I received a call back to Berlin, this time as full professor of physical chemistry and successor of Horst Witt at the TU Berlin in the city center, a very attractive position that I gladly accepted.

■ PROFESSOR AND CHAIR OF THE MAX VOLMER INSTITUTE AT THE TU BERLIN (1991−2001)

The institute in Berlin was very well equipped with personnel, assistant and technician positions, and I was offered enough money to buy and build up three EPR instruments, this time also a pulse X band EPR setup. The three doctoral students from Stuttgart came with me to Berlin and finally all finished their degrees with excellent grades. I also hired Robert Bittl, who came from Gert Kothe's group in Stuttgart as a postdoc; he later became an assistant professor at the TU Berlin. A permanent scientific staff position was given to Friedhelm Lendzian with whom I had done many experiments in the past in the laboratory of Klaus Möbius at the FU. I also joined the Sfb 312 again and in 1993 even became vice chair of this Sfb. After the Sfb 312 was running out after 15 years of funding, I was asked by several scientists in Berlin to start a new Collaborative Research Center. In 1999, we presented a novel grant proposal to the DFG entitled "Protein Cofactor Interactions in Biological Processes" with a fresh crew. The proposal was granted and the program started in 2000 (Sfb 498), when I had already decided to move to the Max Planck Society.

During the years at the TU Berlin, our activities were expanded in many ways. Time-resolved EPR techniques (transient and pulsed) opened new vistas, and we started a closer collaboration with Dietmar Stehlik within the Sfb. New techniques like out-of-phase ESEEM were introduced, and some great experiments were done—mostly due to the excellent work and impact of Robert Bittl (reviewed in ref 182). We also had access to modern high field instrumentation (95 GHz, 360 GHz) via our ongoing collaboration with Klaus Möbius.[70,82] We were the first group to obtain a commercial W band EPR instrument from Bruker and joined the DFG priority program "High Field EPR in Biology, Chemistry and Physics". During the 1990s, we had many visitors who came to give lectures and/or perform experiments with us. The EPR laboratories at the TU were run by Friedhelm Lendzian; he

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did this very well. Much of our scientific success has been due to his profound knowledge, both with respect to instrumentation and sample preparation and handling, his organizational talent, and his continuous efforts to help doctoral students and scientific guests. Together with Dieter Ziessow, we organized the Joint 29th Congress AMPERE/13th ISMAR Conference in 1998 at the TU Berlin—without professional help—with over 700 participants from more than 40 countries. This has been a great success for us and the magnetic resonance community. When I left Berlin in 2000, I offered Friedhelm a permanent contract as group leader at the Max Planck Institute in Mülheim, but he decided to stay in Berlin, which I regretted very much.

The systems studied by us were radicals, radical ions, radical pairs, and triplet states, mostly in photosynthetic systems (reviewed in ref 182). Initiated by the groups of Renger and Witt, we also started to investigate the water oxidizing complex. This was a difficult topic for spectroscopy due to the complexity of the tetranuclear manganese cluster with its at this time unknown structure. I had some experience studying transition metal complexes with EPR/ENDOR techniques from joint projects with a good friend from Klaus Möbius' group, Chris Winscom,[18,42,43,164] but the manganese cluster in photosynthesis presented a much higher level of complexity. We were looking for simpler exchange-coupled systems and started to investigate other metalloproteins like dimanganese catalase. From an old friend, Melvin Klein (UC Berkeley), who happened to be in Berlin on sabbatical, I got the advice to contact Karl Wieghardt at the Ruhr University of Bochum and ask him for his dimanganese model complexes. We started a very fruitful and enjoyable collaboration with Karl supported by the DFG Priority Program "Bioinorganic Chemistry". Due to Karl's profound knowledge of inorganic and also physical chemistry, we were able to gain novel structural insight into mixed valence dinuclear Mn complexes in different oxidation states using multifrequency EPR and ENDOR (reviewed in ref 231). This was backed up with theoretical calculations of the interaction parameters that we developed with Lou Noodleman (Scripps, La Jolla) and Frank Neese (Mülheim).[208]

During this decade, the first single crystals of PS I and PS II from the thermophilic cyanobacterium *Synechococcus elongatus* became available due the continuing excellent work of the group of Horst Witt in our institute, who had developed great expertise in crystallizing membrane proteins. I am still glad that we have made it possible for him to continue his work after emeritus status, and also asked him to join the new Sfb 498. He had two projects on crystallization of PS I and PS II in the Sfb, and already in 2001, the first crystal structures of both photosystems were published in collaboration with Wolfram Saenger from the FU Berlin. My group profited from the availability of the single crystals and the early insight into these exciting structures and published several papers with H. T. Witt and his group members (see, e.g., refs 162 and 166) using expertise developed earlier for single crystals of the bacterial RC.[72]

At the TU Berlin, we also continued our protein preparations and started to do site-directed mutagenesis, not only on bacterial reaction centers but also on the plant systems using *Chlamydomonas rheinhardtii* as the organism. The laboratory was first run by Matthias Kuhn and then taken over by Heike Witt with good success. One of the most successful projects has been the selective exchange of amino acids in the surrounding of the primary donor in bacterial RCs, thereby altering the hydrogen bonds and other interactions of the BChl dimer. We could show why Nature has formed a dimer and how its structure has been engineered for optimum function via interactions with the protein environment. This was performed together with Jim Allen and JoAnn Williams from ASU in Tempe, USA (summarized in ref 182). Similar experiments were later also done by us on the plant photosystems.

At the occasion of one of my frequent visits to UCSD in the mid-1990s, I mentioned that we were interested in the enzyme hydrogenase and had done some work on it. Herb Axelrod, a postdoc of George Feher, told me that one of his friends in Japan is working on [NiFe] hydrogenase and has obtained first single crystals. Through this connection, I got to know Yoshiki Higuchi (Kyoto, now Hyogo University). We started a collaboration that is still continuing and published several papers together, in particular using EPR and ENDOR on hydrogenase single crystals (see, e.g., ref 192). Hideaki Ogata, a former PhD student of Yoshiki, joined my lab and has now been with us for more than 10 years. He is an excellent crystallographer and has done some great work with us.[311,399] It was mainly due to this relation that I got to know Japan, the people, and the culture very well and visited the country several times. Next to the wonderful cities of Kyoto, Tokyo, Nara, Nikko, and Kamakura, our recent visit to the coast near Fukushima shortly after the devastating tsunami and the visit of the city of Hiroshima left a lasting impression on me and my wife and many thoughts concerning the risky use of nuclear power and piling up of nuclear weapons.

Berlin has been an attractive place—both scientifically and culturally—and we had many visitors. My family and I thoroughly enjoyed the sabbatical stay of Richard Cogdell (Glasgow, Scotland), his wife Barbara, and daughter Lucy, who came for one year in 1996 to 1997 with a Humboldt Research Award. Our close friendship is continuing until today. Richard is a fantastic teacher and great scientist, and we owe him the first crystal structure of a bacterial photosynthetic antenna, on which we have also done some joint work related to triplet formation and carotenoids.[155,163] Lucy went to the same school as our son Felix. Now they are both grown up and work as physicians in a clinic—Lucy in Scotland and Felix in Germany. One of Richard's students, Alastair Gardiner, also worked in our group in Berlin as a postdoc with an Alexander von Humboldt Fellowship; he is now in Richard's lab back in Scotland. Alastair was the first Humboldt postdoctoral fellow in our group, and there were many to follow.

Many of the doctoral students in my group were truly excellent and received prizes for their dissertations (Stephan Zech, Monika Fahnenschmidt, Matthias Stein, Hanno Käß). During my time at the TU Berlin, three senior collaborators finished their habilitation: Robert Bittl, who is now full professor of physics at FU Berlin (successor of Klaus Möbius); Petra Fromme, who moved to Arizona State University (Tempe, AZ, USA) as a professor of chemistry; and Johannes Messinger, who first obtained a position with me as group leader at the Max Planck Institute in Mülheim and is now chemistry professor at the University of Umeå, Sweden.

DIRECTOR AT MAX PLANCK INSTITUTE IN MÜLHEIM/RUHR (2000–PRESENT)

In 1999, I received an offer from the Max Planck Society to become a scientific member of the Society and Director at the "Institute for Radiation Chemistry" in Mülheim/Ruhr as successor of Kurt Schaffner. Kurt was doing photochemistry

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and photobiology in his group and had many co-workers in this field. The second director was Karl Wieghardt, a synthetic inorganic chemist who had moved to Mülheim from Bochum a few years earlier and with whom we already had a successful collaboration. After negotiations with the president of the Max Planck Society, Hubert Markl, in Munich, I accepted the excellent offer. For one year, I was holding both positions in Berlin and Mülheim and finally moved in 2001, when the reconstruction of the institute was almost finished and first instruments were delivered. Shortly after this, our apartment near the institute became ready and my wife joined me in Mülheim. In the same year, I was also offered an Honorary Professorship at the Heinrich-Heine University of Düsseldorf. During the first years in Mülheim, I received a lot of advice and direct help from Kurt Schaffner and Karl Wieghardt, which made the beginning in the new surrounding very pleasant.

My move to Mülheim marked the end of radiation chemistry in the institute, and early in 2003, Karl Wieghardt and I founded the "Max Planck Institute for Bioinorganic Chemistry". I have been managing director of this institute from 2004 to 2011. Due to the excellent research done, it soon became well-known in Germany and abroad. Unfortunately, the Max Planck Society was not in favor of a continuation after the retirement of Karl Wieghardt (2010) and the institute's name was changed again in 2012 to "Max Planck Institute for Chemical Energy Conversion". Since we have been working on energy-related topics (water splitting and hydrogen production/consumption in nature), this change has not affected the scientific work in my "Department for Biophysical Chemistry". In the future, we will expand the institute, including the construction of additional buildings, to make room for four instead of two departments, and for new instrumentation. Our Managing Director, Robert Schlögl (2011–present), is working relentlessly paving the way for this to happen. My colleague Frank Neese, who joined the institute as director in 2011, and I are very grateful to Robert, who is doing this job extremely well.

The position of a director at a Max Planck Institute opened the possibility for me to expand the spectroscopy and build a laboratory without budget restraints, which would not have been possible at a German university. For instrumentation, we obtained much space in a reconstructed hall, which was formerly used for the accelerators for radiation chemistry. Additional space was available for new offices and specialized laboratories for biochemical preparations and sample preparations. I received a large number of well-trained technicians with permanent contracts and access to the excellent workshops of the institute for fine mechanics/instrument making, electronics, computer science, quartz glassblowing, etc. This turned out to be of crucial importance for the further development of our instrumentation (see ref 244), and in-house preparations. Sufficient money was available to hire doctoral students and postdocs on MPG stipends, and I was assigned eight group leader positions, many on permanent contracts. With several of the former members of Kurt Schaffner's group, I started collaborations, e.g., with Alfred Holzwarth and Wolfgang Gärtner. For the EPR laboratories, I hired Edward Reijerse (University of Nijmegen), which turned out to be an excellent choice. Ed is not only organizing the EPR facility very well but has also contributed much to our research on transition metal complexes and metalloenzymes, in particular [FeFe] hydrogenases.

During the years, I was able to receive additional support from several grants of the DFG, the BMBF, and the EU in different frameworks. In this context, I would like to mention the impact of the European Networks SOLAR-H (2005–2008) and SOLAR-H2 (2008–2012), which were initiated by Stenbjörn Styring, an old personal friend, in Uppsala (Sweden). I am grateful to him and all co-workers in Sweden who helped to build this network of researchers that contributed to "solar fuel" research. With Stenbjörn and many researchers around the world, I share the firm conviction that we have to work on this problem now in order to have technologies available in time to make us independent of fossil fuels and to fight global warming and climate change for the benefit of mankind.[361,368] The mission of our new Max Planck Institute is basically very similar, and the institute is member of many national, European (AMPEA), and worldwide initiatives (SOFI) in the field of renewable energy and artificial photosynthesis. Earlier this year, an international conference series was established on a meeting in Uppsala related to these topics that will alternate with the respective Gordon Reseach Conference "Renewable Energy: Solar Fuels" that I have chaired in 2012. With Uppsala, I also connect the truly outstanding celebration on the occasion of receiving the doctor honoris causa of the university in 2008, which was a wonderful event for me and my family.

A primary target of our work in Mülheim was the understanding of water splitting and oxygen release in oxygenic photosynthesis in Nature, a topic of great importance not only for chemistry but for science and mankind in general. The related group was first led by Johannes Messinger, a former student of Gernot Renger, who came with me from the TU Berlin. Together with an Alexander von Humboldt postdoctoral fellow from Novosibirsk (Leonid Kulik), we performed and analyzed our first ^{55}Mn ENDOR data on two states S_0 and S_2,[251] supporting and expanding earlier work by David Britt (UC Davis), followed by many other measurements on these states. After Johannes left in 2008, Nicholas Cox from Canberra joined our group, an expert in EPR of complicated transition metal complexes. We also joined forces with Frank Neese's Department and started a close collaboration with Dimitrios Pantazis who delivered the theoretical foundation to interpret the complex spectra. Due to the new highly resolved structure of PS II from Shen and Kamiya in Japan (published in *Nature*, 2011), which also showed the structure of the manganese cluster, the data interpretation could now be based on a more solid geometry. We could show structural changes of the cluster,[343] investigated a third state (S_3) with EPR and ENDOR[394] in cooperation with Alain Boussac from Saclay, and finally determined oxidation and spin states and the spin coupling for all stable intermediates in the catalytic cycle,[403] which is, however, still debated in the photosynthesis community. Models for binding of the two substrate water molecules could also be developed[346,394] and the function of the calcium ion in the cluster proposed.[345] At present, we are close to a basic understanding of the water oxidation cycle in photosynthesis. This success has only been possible due to the continuing support of the Max Planck Society and the great working surrounding in the institute, and last but not least the excellent young scientists who mutually worked on the project, in particular Nicholas Cox and Dimitrios Pantazis during the last years. Both of them received the Ernst-Haage Prize of our institute for their excellent work, and Nick Cox was awarded the Young Investigator Award of the International EPR/ESR Society in 2014. He was actually the fifth young scientist from our group who received this prize (after Robert Bittl 1997,

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Stephan Zech 2003, Leonid Kulik 2007, and Alexey Silakov 2011).

The second line of projects concerns the hydrogenases, enzymes that convert or produce molecular hydrogen. This topic had also been started in Berlin with several engaged students in the lab (Christof Geßner, Marc Brecht, and Stefanie Foerster) and well supported by theory[211] (Matthias Stein). In Mülheim, Maurice van Gastel was working successfully on the [NiFe] hydrogenases, and we also began to study [FeFe] hydrogenases with Ed Reijerse and a very good student from Kazan, Alexey Silakov. Many more smart doctoral students worked on the projects over the years, and the results were summarized in 2007[253] in a special issue of *Chem. Rev.* on "Hydrogen" edited by Bill Tumas (Los Alamos) and me, and again reviewed in 2014, also in *Chem. Rev.*[385] During the past decade, we had recognized that EPR alone cannot solve all the questions and started to use other methods in-house or with cooperation partners. The techniques encompass protein X-ray crystallography (Hideaki Ogata), resonance Raman (Hannah Shafaat), X-ray absorption and emission spectroscopies (XAS and XES, Serena DeBeer), nuclear resonance vibrational spectroscopy (NRVS, cooperation with Stephen Cramer, UC Davis), Mößbauer spectroscopy (Eckhard Bill), FTIR spectroscopy, surface enhanced infrared absorption (SEIRA), (spectro)electrochemistry and protein film electrochemistry (PFE, Olaf Rüdiger), and NMR (Sigrun Rumpel). This broad repertoire of techniques opened the door for obtaining many new data, relevant for understanding hydrogenases and related model systems—not to forget the impact of QC from the group of Frank Neese in our institute.[365,370,417]

A highlight of recent years was the investigation of an oxygen-tolerant [NiFe] hydrogenase[306,323] performed by an excellent doctoral student from Greece, Maria-Eirini Pandelia, in cooperation with the group of Marie Thérèse Giudichi-Orticoni from Marseille. For her outstanding work, Maria obtained the Otto Hahn Medal of the Max Planck Society; she is now starting her own group in the USA. This work laid the foundation for an understanding of the oxygen tolerance of these and related species. The high resolution crystal structure of a [NiFe] hydrogenase obtained by Ogata and Nishikawa[399] showed that hydrogens can be detected by this technique even near the active metal site and for very large systems, which is of general importance for protein crystallography far beyond the specific case of hydrogenases. Koji Nishikawa is now back in Japan and received a position as Assistant Professor in Hyogo. The demonstration that hydrogenases can be stabilized in a redox polymer, which protects them from oxygen damage and high potential deactivation at the electrode, is important for possible biotechnological applications of hydrogenases.[391,411] This work has been done by Olaf Rüdiger in cooperation with Wolfgang Schuhmann's group from Bochum. Through a close collaboration with Thomas Happe (Bochum), Marc Fontecave (Grenoble/Paris), and Vincent Artero (Grenoble), we could show that chemically synthesized precursors of the active site of [FeFe] hydrogenases could be inserted into the apoprotein, yielding fully active enzyme.[367,375] This procedure opened the possibility to create new hybrid enzymes with altered and possibly improved properties;[405] furthermore, it allows one to specifically label the site with markers[420] or to obtain a better picture of the electronic structure.[416] The spectroscopic work on these systems has all been done by Agnieszka Adamska-Venkatesh and the preparations by Judith Siebel, two excellent PhD students of my department. This work together with earlier data allowed us to set up the reaction mechanism of the catalytic hydrogenases, reviewed in ref 385. The investigations of the native hydrogenases have been supplemented by model system studies; these were either obtained from external collaborators, Marcetta Darensbourg (Texas A&M, College Station),[348,389] Thomas Rauchfuss (Urbana, IL),[349,416,419] or Sascha Ott (Uppsala, Sweden),[318,378] or synthesized in our group by an excellent doctoral student, Katharina Weber.[353,390,415]

Much of our early work has been done together with Siem Albracht (Amsterdam), which was a very fruitful collaboration. We also worked on other hydrogenases that came mainly from *Ralstonia eutropha* prepared in the group of Bärbel Friedrich and Oliver Lenz at the Humboldt University Berlin, both long-term cooperation partners. Recently, the regulatory hydrogenase (RH) was spectroscopically characterized,[413] the system on which we first observed the hydride bonded in the Ni−Fe bridge.[204]

For many years in the 1990s, we enjoyed a close collaboration with the group of Astrid Gräslund (Stockholm) on ribonucleotide reductase. This has recently been renewed with joint studies on some heterodinuclear (FeMn) metalloproteins.[372,395] It was Astrid (Secretary of the Nobel Prize Committee for Chemistry in Stockholm) who asked me to join the Council of the Lindau Nobel Laureate Meetings. In 2004, I was elected as member of the Council. Since then, the two of us have organized the chemistry meetings every three years and also helped in the meetings on physics and physiology/medicine. In recent years, the Lindau Conferences have become very popular. For the interdisciplinary meeting in 2015, we had 65 Laureates present and more than 650 young researchers from over 80 nations. The young scientists had been selected by the council members from an even larger group of recommended students. The Lindau Nobel Laureate Meetings are bringing together the best young researchers from all over the world to personally meet those scientists who have been honored by the Noble Prize for their outstanding work. For me, it is very enjoyable to be part of this event every year in one of the nicest places in Germany and meet so many exciting and inspiring people.

In 2009, we invited Professor Klaus Möbius (Physics Department, FU Berlin) to join our institute in Mülheim as a permanent guest scientist. He accepted the offer and brought his home-built 94 GHz EPR/ENDOR/ELDOR spectrometer with him from the FU Berlin. At the same time, we hired Anton Savitsky from his former lab as a group leader. Since this time, Klaus and Anton have been working together on paramagnetic species and problems related to the mission of the institute together with several guest scientists, e.g., Yuri Grishin (Novosibirsk), Alexey Semenov (Moscow), and Giovanni Venturoli (Bologna). It has been a very fruitful and enjoyable cooperation and led to more than 20 scientific papers (see, e.g., refs 377, 400, and 406). Even more important is the great help in instrumentation development and improvement that we obtained from this connection.[359,376] Many of our recent more difficult experiments would not have been possible without this support. On the occasion of Klaus' 75th birthday, we organized an EPR Symposium in his honor to which a large number of scientists and friends came to Mülheim. I am very grateful to Klaus for all his personal help, his continuing scientific support, and a wonderful friendship—also with his family. Klaus is one of the guest editors of this special issue together with Brian Hoffman (Northwestern University, Illinois). Brian has always

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been a role model for us who has contributed so much to understand complex metalloproteins, metal complexes, and radicals using magnetic resonance techniques. I want to thank both of the Guest Editors for all the work they have invested in this special issue and all the colleagues and friends who contributed to it.

SOME PERSONAL WORDS AND THANKS

As is obvious from my CV, I have always been working at the interface of two or even more scientific disciplines, which was not always a simple task. Cross-disciplinary work requires special efforts, but I believe that the outcome is particularly satisfying and it is a simple truth that the most exciting things in science often happen at the interface of the classical disciplines. The Sfbs ("Sonderforschungsbereiche")—the best funding structures of the German Science Foundation (DFG)—are entirely based on the idea of cross-disciplinary work, and I have been working in Sfbs almost through my entire scientific career. However, there is a caveat to all of this; at least for me, it has been difficult to find my place in the traditional academic landscape of German universities, and I am therefore quite happy to end my scientific career working for the Max Planck Society where the institutes are structured in a different way.

The results and achievements of my laboratory would not have been possible without the efforts of the members of my research group. I sincerely hope that all of them have enjoyed their work as much as I did. It has been a privilege to work together with so many gifted young graduate and postgraduate students that make you feel young, at least at heart, as George Feher expressed it beautifully in his article published on the occasion of my 60th birthday (*Appl. Magn. Reson.*, 2010, special issue edited by Klaus Möbius, Kev Salikhov). I also hope that the students have successfully used the acquired knowledge and skills for their own careers. I also want to thank our many collaborators from other institutions around the world for their input and effort to make our projects a success. It was very nice both scientifically and personally to work together with so many scientists from different nations, which gave me, my group, and my family the opportunity to get to know many foreign countries, their cultures, their traditions, and their current and past history. The exchange of students and scientist is a great means to bring together young people and is the best way to foster international understanding. Last but not least, I also want to thank all the other members of the institute in Mülheim who are successfully covering my back, helping wherever they can to make my work here as pleasant as possible.

I have been gifted with a wonderful family. My wife Gisela and I have been married now for 36 years, and we have known each other even much longer. We are proud that our son Felix made his medical doctor; he is working in a large clinic in a responsible position and will soon finish his specification as cardiologist. To our pleasure, he also got married earlier this year. The support of my family has been extremely important for me and my career. Without my wife, her constant help at home and her great support of my professional work in Mülheim, it would not have been possible for me to so efficiently perform at the institute. I owe her a lot for doing this for me. She knows that I love her very much—but also my scientific work.

Wolfgang Lubitz

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