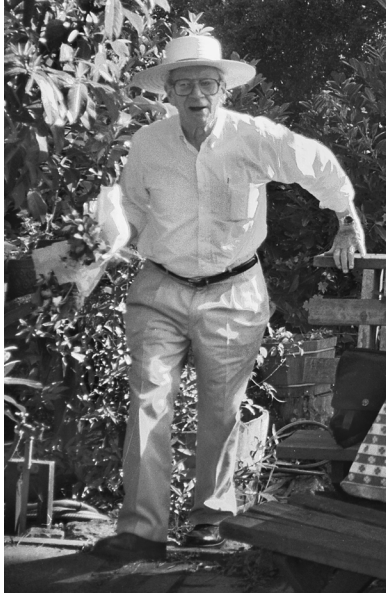


Friday, February 16, 2001

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[**Note to the publisher:** The footnote is currently at the end of the article; it should appear as a footnote on printed page 1: Govindjee]

Obituary: Remembering Melvin P. Klein[‡]



Mel Klein (1921-2000)

Mel Klein died suddenly, at the age of 78, at his home in Berkeley on May 28, 2000, as he was preparing to go bicycling in Golden Gate Park in San Francisco. Mel was born on July 27, 1921, in Denver, Colorado, to an American mother and a Hungarian father, and he attended, appropriately for half a Hungarian, the Teller Elementary School. As a young boy, Mel displayed an early affinity for biology and physics, playing endlessly with both plants and baseballs. He went on to Aaron Gove Junior High where, by age 10, it had become clear that he was a prodigy with gadgets and electronics. He became a radio ham at age 11, infecting his teacher with this affliction and founding a school radio club. He stayed an avid ham radio enthusiast till his death, serving as radio liaison on the Berkeley, California, hills fire patrol. It is told that young Mel, the gadgeteer, stalked the residents of his Denver neighborhood, confronting them in the streets, challenging them to give him a broken radio to fix. At East Denver High School, Mel was an outstanding student and a much decorated scout leader. He attended Denver University, working

at the same time as an "engineer" in local radio stations. December 7, 1941 came, he quickly found a job with the Army Air Corps at Logan Utah air base as a civilian radio engineer. Relocating to a subsidiary air base outpost at Las Vegas, Nevada, Mel did a lot of flying in C-47s with outside temperatures of -60°C or lower. He later got a job with the Office of War information in San Francisco as an engineer. After a year in which he learned to love the San Francisco bay area, he went to Washington, D.C. to a firm of consulting radio engineers.

Mel's early love of radio frequency (rf) technology as a ham radio buff, his affection for radio techniques and his formal pre-war engineering training at Denver University led him into radar research at the Harvard Radio Research Lab that worked on countermeasures to the Massachusetts Institute of Technology (MIT in Boston/Cambridge, Massachusetts) Radiation Lab's development of radar. This was followed by a stint in the Pacific theater, where he was attached to a group of civilian scientists/technicians who were part of an intelligence section of General MacArthur's headquarters working with radio antennae, following him closely from Australia to New Guinea to the Philippines. During that period, Mel learned physical optics and decided to learn more about physics and electromagnetic theory.

These wartime radio technologies led directly to the rapid advent of magnetic resonance spectroscopies after the war. In 1945, back in Cambridge, Mel listened to Felix Bloch talk about an idea he had for doing "something magnetic." In fact, Bloch essentially offered Mel a graduate student fellowship at Stanford University to work on this "thing." Mel remembered that he was not yet turned on to the idea of graduate school and hence, he reminisced, what became Bloch, Hansen and Packard could have been Bloch, Hansen and Klein, placing Mel at the epicenter for Nuclear Magnetic Resonance (NMR) development had he chosen that path.

Deciding to return to school, Mel did the first year of Medical School, and then switched to Physics and Biophysics. Mel went to Dave Sloan's lab at the University of California (UC) at Berkeley, where George Feher was also working at that time. He finished the degree in Physics in 1952, joined the UC Radiation Laboratory in 1952, and obtained his Ph. D. degree in Biophysics in 1958. During this period, Mel became acquainted with the Luis Alvarez group and was part of the radiation-monitoring group that was present at the 'Mike shot' at Eniwetok atoll. Mel spent some time with Alvarez at Livermore (California) where he was involved with the rf for a giant proton LINAC (linear accelerator) that quickly led him into the world of magnetic resonance. He learned about NMR as a way of measuring magnetic fields for a mass spectrometer being used

for the measurement of Li isotope ratios. Looking at a table of spins and moments, Mel realized that NMR could also be used to directly obtain this ratio. Management concurred and bought the first Varian instrument for delivery to Livermore; this led to a number of papers describing the fundamental magnetic properties of various nuclei. In these early years of his research career, Mel worked on NMR experiments with collaborators such as John Waugh and Walter Knight. He also started his first Electron Paramagnetic Resonance (EPR) experiments, initially working with Ray Ward at Livermore. Mel also collaborated with Alan Portis's group in designing an early superheterodyne EPR spectrometer, including a novel bimodal cavity that inspired many of his subsequent students as one of the star items in Mel's amazing collection of microwave and rf components.

In 1960, Bill Blumberg, one of Erwin Hahn's students who had gone to Bell Labs invited Mel to come for a year. The Bell Labs were a super place and many friendships were forged. There he worked mostly on paramagnetic solids, and the results of collaborations and close friendships with scientists such as Bill Blumberg, Bill Mims, Stanley Geschwind, and Jack Peisach echoed throughout Mel's subsequent career. It was shortly after returning to Livermore, in 1961, that George Barton and Mel came up with the idea of digital signal averaging methods for noise reduction from which we have all benefited so immensely. Lawrence Berkeley National Laboratory (LBNL) Director Charles Shank said in his tribute to Mel that "the digital signal-averaging technique has become the *sine qua non* of modern Fourier transform NMR spectroscopy and has permeated most fields of experimental science."

In 1963 Melvin Calvin invited Mel to join his group at UC Berkeley to help strengthen the staff of the new Round House. Mel became involved in an extraordinary range of activities, making important contributions to NMR, EPR, Mössbauer, XPS (X-ray photoelectron spectroscopy), perturbed angular correlation, MCD (Magnetic Circular Dichroism), and microwave photoconductivity. Except for brief sabbaticals in Paris (1966-67) on a Guggenheim Fellowship and in Berlin (1988-89) on a Humboldt Award, Mel spent the rest of his biophysics career at UC Berkeley. He carried out a broad and vigorous program of magnetic resonance spectroscopy in physics, chemistry, and biology. His work over these years continued to include fundamental instrumentation development, including the building from the ground up of a number of NMR and EPR spectrometers. Also, in the late 60s, Mel obtained one of Varian's first NMR instruments with a superconducting magnet, a 220 MHz proton Continuous Wave

spectrometer, and quickly converted it into an Fourier Transform instrument, one of the few around at that time and the first in an academic institution.

The array of scientific topics to which Mel brought his magnetic resonance expertise is comprehensive indeed, including photosynthetic systems, ferrichrome, porphyrins, paramagnetic proteins, excited spin states, radioactive nuclei, small organic chemical species, liquid crystals and biological membranes, solid state systems, *in vivo* nuclear spins, mechanisms and applications of Chemically Induced Dynamic Nuclear Polarization (CIDNP), and coherent Raman beat EPR spectroscopy. In recognition of Mel's contributions to magnetic resonance spectroscopy, he was elected as a Fellow of the International EPR Society.

In the early 70s, Mel was turned on by the potential of spectroscopy using Synchrotron Radiation, just then becoming available at the Stanford electron storage ring, to study the Mn complex involved in photosynthetic oxygen evolution. In a review article he wrote about photosynthetic oxygen evolution for a special issue of Photosynthesis Research dedicated to the memory of Bessel Kok, Mel Klein reminisced on how he was initiated into the realm of photosynthesis research, especially with regard to the Mn complex in the Oxygen Evolving Complex(OEC) (Klein et al. 1993):

"This article reflects the personal perspectives of one of us (MPK) who came to the problem without significant previous experience in photosynthesis research even while hearing about it on an almost daily basis because of the foment in the Melvin Calvin Laboratory. In the late 1960s, one of us (Ken Sauer) was aware of the experiments on the flash induced oxygen evolution by (Pierre) Joliot and coworkers, and the various interpretations by him and by Kok. One of Sauer's postdoctoral fellows, Dr. Charles Weiss, undertook to extend these experiments using a Q-switched ruby laser, just then becoming available. It was in that context that Klein became aware of the problem.

It was also during this period that Sauer's students G. T. Babcock and R. E. Blankenship were performing EPR experiments that showed that, upon treatment with Tris, thylakoid membranes released EPR detectable Mn into the inner (luminal) aqueous spaces with concomitant loss of oxygen evolution. Following rewashing in a Tris-free buffer containing DCIP/ascorbate, the Mn(II) EPR signal became undetectable and oxygen evolution was restored.

In late 1972 or early 1973, Professor Marvin Cohen of the Berkeley Physics department who was chairman of the Gordon Research Conference on "The Physics and Chemistry of

Solids: Deep Level Spectroscopy," invited Klein to attend. The latter, claiming that such topics were far from Biophysics, acquiesced to Cohen's blandishments: "Come! You'll learn something."

It was during that Conference that Dr. Dale Sayers lectured on Extended X-ray Absorption Fine Structure (EXAFS). It became immediately apparent that such studies could be used to probe the structure of the Mn moieties and address the question of whether the oxidation state(s) of the Mn were influenced by light. It was known that the binding energies of core electrons reflected the oxidation states of elements. Such information was being derived at that time from X-ray photoelectron spectroscopy, XPS.

Fortunately, members of the physics community recognized that to make the experiments of Sayers, Lytle and Stern accessible to a larger community, it would require a convenient source of intense, collimated and tunable X-ray photons. They were our colleagues at Stanford who had the insight to begin the conversion of the Stanford Positron Electron Accelerator Ring, SPEAR, into a synchrotron radiation source. Using only the X-ray flux emanating from a conventional X-ray tube, it would have been impossible to perform experiments on samples as dilute in Mn as are thylakoid membranes. Also, X-ray absorption spectra from such dilute biological samples could not be collected in the absorption mode, and to overcome this problem our group introduced the fluorescence method of collecting such spectra. We demonstrated that the X-ray absorption spectrum of Mn in a leaf could be collected using such a method.

It was with this background that our group first began study of the Mn atoms in PS II."

Mel's contributions to X-ray Absorption Spectroscopy (XAS) have been marked by his interests in applying state-of-the-art technology, an example of which was the introduction of the fluorescence detection system. The X-ray detectors at this time used scintillator coupled photomultiplier tubes. Mel realized that, for the detection of dilute samples, it would be advantageous to use solid-state energy-discriminating detectors. However the linearity of shaping amplifiers in use at that time was too limited. So Mel provoked the detector group in LBNL to design and build faster shaping amplifiers. It was with such a detector that some of the most important contributions to the X-ray absorption spectroscopy of Mn in the OEC were obtained. He and his coworkers have made seminal contributions to what is known about the structure and oxidation states of the Mn complex as it cycles through its five light-induced states. At the time of his death, Mel was actively pursuing X-ray spectroscopy research of the Oxygen Evolving

Complex (OEC) in plants, a field to which he had made fundamental contributions for the past quarter century. Mel also was instrumental in establishing biological research facilities at Berkeley Lab's recently built Advanced Light Source.

In the 1980's, as his manganese EXAFS program grew into a major part of Mel's overall research program, the natural tie with Mn EPR spectroscopy developed alongside. Interestingly, Mel's interest in Mn in plants seems to have started during his sabbatical at Bell labs. He relished relating a story about how, while driving with Bill Blumberg to Bell Labs, they stopped on the roadside and picked some green leaves, which they ground up and stuck in the EPR to observe a characteristic six-line Mn^{2+} signal. This experiment with a leaf was later repeated by obtaining a X-ray absorption spectrum of Mn, as described above.

Mel's EPR contributions to oxygen evolution research include: the initial EPR characterization of mixed valence dinuclear Mn clusters, studies of chemical exchange and magnetic isotope effects on the multiline EPR signal lineshapes, EPR characterization of thermophilic cyanobacterial PS II, pulsed EPR applied to the paramagnetic PS II states, parallel mode EPR of PS II and Mn model systems, orientation effects on ammonia-treated and native PS II membranes, and investigations of the EPR properties of the S_0 state. There was always a great natural synergy between the EXAFS and EPR efforts, for example with careful EPR analysis of the S-state advancement of EXAFS samples, or of the alignment of oriented membranes, or in the use of EPR in defining interesting EXAFS experiments based on known EPR effects, such as the formation of the $g=4.1$ signal or alteration of the multiline EPR signal with Sr^{2+} or NH_3 treatment.

In addition to his substantial intellectual contributions to fundamental science and research, Mel Klein was a mentor and good friend to a large number of students, postdoctoral researchers and colleagues. Every day working with Mel brought a dozen new ideas. After a few months, a new student learned that she/he didn't have to actually follow up on all of them! In fact, many of Mel's suggestions often didn't really clearly register at first, but might only achieve clarity after a few days of rumination! Mel's favorite way of describing his approach to students was as he described it, "I sent them fishing with the hope that they would get caught." A great delight was spending time with Mel rummaging through the equipment drawers and closets of the Calvin lab. Mel knew what every piece of equipment had been used for, and would share fond memories about the experiments and colleagues. His office door characteristically stood

open, and he was never too busy to discuss significant science with anyone from undergraduates to senior colleagues. His visitors typically departed with new insights and clever ideas as rewards of conversations with Mel. This quality, together with the respect he showed everyone, whether student, administrator, or scientist, earned him universal admiration and affection. He was particularly valued for his expert and authoritative advice on the broadest spectrum of physics, biology and chemistry. He loved to design and build novel instrumentation, and he was often at the console showing students how to tweak the controls to optimize performance. His hands were as gifted as his mind. Mel was always very generous about taking his students to meetings, and he would make sure they made acquaintances with his many friends. Many important scientific connections were fostered by Mel in this way. It's another way his spirit lives on today.

Mel's students and associates over the years honored him with a symposium held in January 1998. In a Festschrift issue of the *Journal of Physical Chemistry B* issued on October 15, 1998, the symposium proceedings were accompanied by a biographical sketch, a list of over 200 of his publications and an even longer list of scientific associates with whom he interacted during his long career. On the occasion of the symposium honoring Mel, his friend and colleague Alex Pines concluded his talk with the following observation "Mel Klein has educated generations of students and postdocs, many of whom have gone on to stellar careers, and he has been and remains an enduring inspiration to us all. But Mel also has that rarest of human qualities - he is a mensch, a real mensch. Dear Mel, I am sure that I write on behalf of all your students and colleagues when I say that you have earned our love and our respect. All the bypasses in the world cannot change the integrity and the warmth of your heart."

In announcing Mel's death, Graham Fleming, director of the Physical Biosciences Division of LBNL, of which Mel was a member, wrote "Mel embodied the spirit of curiosity and interest in the world around him that marks a true scientist and an intellectual. To say he will be terribly missed is an insufficient expression of our loss."

Mel Klein's major contribution to photosynthesis research was his dedicated pursuit of the structure of the Mn complex involved in photosynthetic oxygen evolution, and the mechanism by which the Mn complex catalyzes the oxidation of water to oxygen. EPR and X-ray absorption spectroscopies have provided much of the information that is now known about the structure of the Mn complex, and Mel's research on this topic was characterized by the use of both of these techniques and the elegant interplay between these two methods. Selected

publications of Mel related to photosynthesis, followed by a list of people who interacted with Mel during the course of their research in photosynthesis, are included below. The impact of Mel's work is the basis of research going on in dozens of laboratories throughout the world. Those of us who worked most closely with him cherish the memories of great science and good times together with an unforgettable human being.

Selected Publications in Photosynthesis by Mel Klein

- Britt RD and Klein MP (1992) Electron Spin Echo Envelope Modulation Studies of Mixed Valence Manganese Complexes: Applications to the Catalytic Manganese Cluster of Photosynthetic Oxygen Evolution. In: Bagguley D (ed) Pulsed Magnetic Resonance: NMR, ESR and Optics: a recognition of Erwin L. Hahn, pp 361-376. Clarendon Press, Oxford
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- Burghaus O and Klein MP (1995) Coherent Raman Beat and ESEEM EPR Spectroscopy Applied to a Transition Metal Complex: Comparisons of Sensitivity and Spectral Resolution. *Chem Phys Lett* 243: 480-485
- Cinco RM, Robblee JH, Sauer K, Yachandra VK and Klein MP (1998) Strontium EXAFS Shows Calcium Cofactor Proximity to the Manganese Cluster in Oxygen-Evolving Photosystem II. *J Phys Chem B* 102: 8257-8265.[This was a Festschrift Issue of the Journal of Physical Chemistry B for Mel Klein and Ken Sauer]
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- Kim DH, Britt RD, Klein MP and Sauer K (1990) The g=4.1 EPR Signal of the S₂ State of the Photosynthetic Oxygen Evolving Complex Arises from a Multinuclear Mn Cluster. *J Am Chem Soc* 112: 9389-9391
- Kirby JA, Robertson AS, Smith JP, Cooper SR and M.P. Klein MP (1981) The State of Manganese in the Photosynthetic Apparatus I. EXAFS Studies on Chloroplasts and Di- μ -oxo Bridged di-Manganese Compounds. *J Am Chem Soc* 103: 5529-5537
- Klein MP, Sauer K and Yachandra VK (1993) Perspectives on the Structure of the Photosynthetic Oxygen Evolving Mn Complex and its Relation to the Kok Cycle. *Photosynthesis Research*. 38: 265-277
- Latimer MJ, DeRose VJ, Mukerji I, Yachandra VK, Sauer K and Klein MP (1995) Evidence for a Calcium Binding Site in Close Proximity to the Manganese Cluster of the Oxygen Evolving Complex: EXAFS Studies of Strontium-substituted Photosystem II Membranes. *Biochemistry* 34: 10898-10909
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Yachandra VK, Sauer K and Klein MP (1996) Manganese Cluster in Photosynthesis: Where Plants Oxidize Water to Dioxygen. In: Holm RH and Solomon EI (eds) *Bioinorganic Enzymology*, *Chem Rev* 96: 2927-2950

List of people involved in Photosynthesis Research with Mel Klein

Students, Postdocs, Collaborators and Sabbatical Visitors, alphabetically arranged below, worked with Mel Klein on some aspect of Photosynthesis. Many in this list were students or postdocs of Ken Sauer, but who nevertheless interacted with Mel in Photosynthesis Research. This is an example of the wonderful synergy that Mel and Ken brought to research on the Mn Complex in PS II in the Calvin Lab.

Joy C. Andrews, William H. Armstrong, Lars-Erik Andréasson, Elodie Anxolabehere-Mallart, Gerald T. Babcock, Emmanuele Bellacchio, Uwe Bergmann, Robert E. Blankenship, Genevieve Blondin, Mike Boska, R. David Britt, Gary W. Brudvig, Melvin Calvin, John L. Casey, George Christou, Roehl M. Cinco, James L. Cole, Stephen P. Cramer, Holger Dau, Victoria J. DeRose, Susan L. Dexheimer, G. Charles Dismukes, Carmen Fernandez, Jean-Jacques Girerd, David B. Goodin, Ronald D. Guiles, John Hearst, Derek Hodgson, Jon A. Kirby, Masami Kusunoki, Matthew J. Latimer, Wenchuan Liang, John L. McCracken, Ann E. McDermott, Karen L. McFarlane, Johannes Messinger, Ishita Mukerji, Gary T. Olsen, Vincent L. Pecoraro, Shelly A. Pizarro, John H. Robblee, Theo A. Roelofs, Alan S. Robertson, Annette F. Rompel, Ken Sauer, Vinita Singh, Dietmar Stehlik, Joseph P. Smith, Sun Un, Henk Visser, Karl Wieghardt, Thomas Wydrzynski, Vittal K. Yachandra, Wa On Yu, Jean-Luc Zimmermann.

R. David Britt

Dept. of Chemistry, Univ. of California, Davis, CA

Kenneth Sauer

Dept. of Chemistry, Univ. of California, Berkeley, and Melvin Calvin
Laboratory, Physical Biosciences Division, Lawrence Berkeley National Laboratory,
Berkeley, CA

Vittal K. Yachandra

Melvin Calvin Laboratory, Physical Biosciences Division,
Lawrence Berkeley National Laboratory, Berkeley, CA

‡ Portions of this tribute are edited excerpts from a talk given in honor of Mel Klein by Alex Pines, which appeared in *Journal of Physical Chemistry B* (1998) 102, 8123-8124, and major parts of this remembrance are reproduced from a tribute to Mel Klein to appear in a special issue of *BBA Bioenergetics Reviews on Photosynthetic Oxygen Evolution* edited by Jonathan Nugent. The present Obituary for "*Photosynthesis Research*" was edited by Govindjee.