



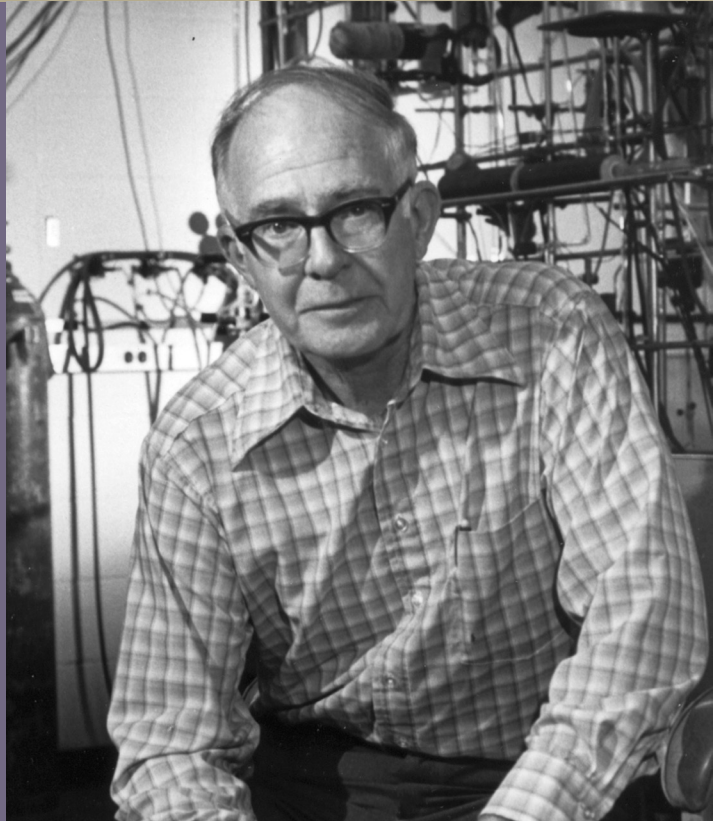
Raymond Davis Jr.
1914–2006

BIOGRAPHICAL

Memoirs

*A Biographical Memoir by
Kenneth Lande*

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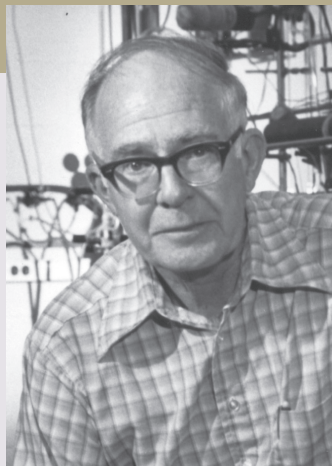
NATIONAL ACADEMY OF SCIENCES

RAYMOND DAVIS JR.

October 14, 1914–May 31, 2006

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Neutrino astronomy¹, the observation of neutrinos from extraterrestrial sources, began in 1966, when Raymond Davis, Jr. turned on his deep-underground chlorine-based neutrino detector. Over the next three decades, the lower-than-predicted solar neutrino flux that Davis observed confused the scientific community. Was our understanding of energy generation in the core of stars flawed? Was there an unforeseen experimental error? Or were neutrinos more mysterious than we had anticipated? The scientific career of the remarkable scientist Raymond Davis played an integral role in unraveling the complex nature of neutrinos and in confirming our nuclear fusion model of energy generation in the core of the Sun.



Raymond Davis Jr.

By Kenneth Lande

Early life and work

The field of observational neutrino astronomy began in 1966 with the initial operation of the chlorine-based solar neutrino detector that Raymond Davis, Jr. built in the Homestake Gold Mine in Lead, South Dakota. The Sun is the brightest neutrino source in the sky, and thus it was the obvious source with which to initiate a new field. For the next two decades, Davis's detector was the only operating astronomical neutrino telescope. The results of these observations were remarkable ⁽¹⁾.

Strangely enough, this experiment was linked to a puzzle that arose a century earlier with Charles Darwin's 1859 publication of *On the Origin of Species*. Kelvin challenged Darwin's conclusion that the Earth was over 300 million years old. On the basis of the Sun's gravitational compression time, Kelvin calculated that the Sun had not radiated energy at its current rate for more than ~ 30 million years—far shorter than Darwin's evolutionary timescale. Of course, when Kelvin challenged these findings in 1860, nuclear reactions were unknown, and thus fusion reactions with their enormous energy release were unimaginable ⁽²⁾.

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How did Davis, a chemist by training, become involved in this problem? The remainder of this section describes Davis's early life and work, including his journey from chemistry to physics.

Raymond Davis, Jr. was born in Washington, D.C., on October 14, 1914. His father, who was self-educated, was a photographer at the National Bureau of Standards and eventually became chief of the Photographic Section. Davis, Sr. encouraged his son's early interest in chemistry and taught him to construct scientific apparatus, which proved critical for the creation of Davis's unique neutrino detector.

Davis began his chemistry studies at the University of Maryland shortly after Wolfgang Pauli, in December 1930, announced his "desperate" solution to the lack of constant total visible energy of the secondaries in beta decay. According to Pauli, there had to be a missing, invisible, neutral zero-mass particle in the final state: the neutrino⁽³⁾. Pauli did not connect this missing particle with energy generation in the Sun. That step came in 1938, the year in which Davis graduated from the University of Maryland and in which Hans Bethe gave his famous lectures on the two possible fusion processes in the Sun—the carbon cycle and *p-p* fusion—at nearby George Washington University⁽⁴⁾. In both of these processes, four protons convert into ⁴He with the emission of two electrons and two neutrinos. Because the mass deficit (binding energy) of ⁴He was well known, the number of neutrinos produced per second was proposed to be simply the energy per second emitted by the Sun divided by the binding energy of ⁴He, multiplied by two. Even though Davis was currently living only a few miles away from where these momentous lectures took place, it would be another decade before he developed an interest in what had occurred there.

Davis briefly worked for Dow Chemical Corporation before enrolling at Yale University, where in 1942 he received a PhD in physical chemistry. Immediately after graduation, Davis entered the U.S. Army and spent most of World War II at the Dugway Proving Grounds in Utah. After the war ended in 1945, Davis took a job with Monsanto Chemical Company and began working on radiochemistry, a then-new field and a critical link to his future scientific activities.

Initial neutrino detector concepts

The following year, in 1946, Bruno Pontecorvo devised a radiochemical detection technique for neutrinos. His classic five-page paper almost completely described the radiochemical technique that Davis was later to employ in his solar neutrino detector.

In his paper, Pontecorvo described how neutrinos interacted with ^{37}Cl to form ^{37}Ar : $\nu_e + ^{37}\text{Cl} \Rightarrow ^{37}\text{Ar} + e^-$. (Note that the nomenclature used here is recent: In 1946 there was only one known neutrino species, and there was as yet no distinction between neutrinos and antineutrinos.) The experimental concept was to remove the ^{37}Ar from the chlorine-containing fluid, concentrate it in a small proportional counter, detect its decays (^{37}Ar was unstable and decayed with a 35-day half-life), and from that information determine the flux of neutrinos. Pontecorvo presented his design for a neutrino detector at a 1946 meeting of the Canadian Physical Society⁽⁶⁾. The U.S. Atomic Energy Commission (USAEC) was concerned that such a detector could be used to locate nuclear submarines, and therefore it classified Pontecorvo's paper. Of course, the power reactors that the USAEC was concerned about emitted antineutrinos, but it would take another few years to demonstrate the distinction between neutrinos and antineutrinos.

There were several differences between Pontecorvo's plan and the one eventually executed by Davis. Pontecorvo had suggested using carbon tetrachloride (CCl_4) as a source of the needed ^{37}Cl atoms. For the underground detector, Davis substituted a safer, chlorine-containing liquid, perchloroethylene (C_2Cl_4). Pontecorvo considered separating the ^{37}Ar from the CCl_4 by boiling the liquid, whereas Davis used a subtler approach, bubbling helium through the liquid and flushing out the argon atoms.

In 1949, Luis Alvarez refined Pontecorvo's Cl-Ar neutrino-detection technique⁽⁷⁾. Alvarez recognized that the Cl-Ar detector might detect only neutrinos and not antineutrinos, but he dismissed this possibility on the basis of a misleading double-beta-decay experimental result. He also described the helium-sweep method of extracting argon atoms from the detector fluid and suggested using a liquid nitrogen-cooled charcoal trap to remove the argon atoms from the helium gas. He even mentioned the use of a Toepler pump to move the argon atoms from the charcoal trap to the proportional counter, a critical noncontaminating gas pump that Davis later used. Using improved cross-section estimates over those employed by Pontecorvo, Alvarez concluded that an appropriate detector for reactor neutrinos required ~ 40 tons of CCl_4 , much more than the 1 ton Pontecorvo anticipated, and that this detector had to be within ~ 10 m of a power reactor to produce a detectable signal. Finally, Alvarez devoted considerable discussion

to background processes that could produce ^{37}Ar and to others that could falsely trigger the ^{37}Ar -decay detector, the miniature proportional counter. There is no indication that Alvarez ever proceeded past this document in trying to build such a neutrino detector. An additional, unexpected result of Alvarez's conclusions regarding realistic detector size, proximity to the reactor, and signal was that the USAEC declassified the 1946 Pontecorvo paper.

The early years at Brookhaven

During this period, Davis was making the transition from industrial chemistry to the world of research. In 1948, he accepted a position in the chemistry department at the recently created Brookhaven National Laboratory. According to Davis, the first thing he did was ask the chemistry department chair, Richard Dodson, what he was to do. Dodson's reply was simple: "You figure it out." So Davis went to the library and found a new review article about neutrinos by H.R. Crane ⁽⁸⁾. He was fascinated by the idea of looking for these ephemeral particles. By 1949, the critical elements had come together—Davis's interest in the idea of detecting neutrinos, Pauli's "ghost" particle, and the writings of Pontecorvo and Alvarez that stimulated Davis's experimental genius. Davis went about translating these ideas and concepts into an operating neutrino detector and into a precise prediction of the energy-dependent flux of solar neutrino emission—the signal.

In 1955, Davis published the first measurements taken with Cl-Ar detectors ⁽⁹⁾. There were two detectors, one with 200 liters of CCl_4 and another with 3900 liters of CCl_4 . In this paper, Davis discussed two motivations: (a) to determine whether antineutrinos emitted by a nuclear reactor have the same interaction as neutrinos, and so drive the $^{37}\text{Cl} \Rightarrow ^{37}\text{Ar}$ transition, and (b) to search for neutrinos coming from the Sun and, in the process, to measure the cosmic ray-induced background.

The 200-liter detector had a high-altitude exposure at the Mt. Evans High Altitude Laboratory (4310 m above sea level, or 610 g cm^{-2} atmospheric depth) and sea-level exposures at Brookhaven both close to and far from the Brookhaven High Flux Research Reactor. The detector observed a positive signal only in the high-altitude exposure. The 3900-liter detector was exposed to the Brookhaven Reactor, then was buried 5.8 m below the surface of the Earth (970 g cm^{-2} additional overburden) to observe the background-reduction effects that additional overburden would produce. A positive signal was observed in the reactor exposure. Davis attributed this signal to the $^{37}\text{Cl} (p, n)^{37}\text{Ar}$ reaction and assumed that the protons originated in cosmic rays. No further tests were carried out to

rule out either reactor antineutrinos or neutrons. In retrospect, it would have been useful to measure the signal rate in the 3900-liter detector at a location on the surface far from the reactor. However, as discussed below, this experiment was repeated at the Savannah River reactor with a considerably higher antineutrino flux.

Davis's 1955 ⁽⁹⁾ paper contains a fairly detailed description of the critical experimental components: the argon-extraction system, the gas-purification system, and the counting system. The counting-system background rate, 0.15–0.25 counts min⁻¹, severely limited the sensitivity of these experiments. Not for another two decades was a truly low counting-system background rate achieved. This process involved (*a*) replacing the Geiger–Müller counters with low-background proportional counters, (*b*) replacing the six anticoincidence counters with a fully enclosing NaI crystal, (*c*) increasing the thickness and purity of the counting shield, (*d*) installing rise time–detection circuitry, and (*e*) placing the entire counting system deep underground. However, the 1955 experiment clearly demonstrated that Davis was able to quantitatively extract and handle samples of only a few ³⁷Ar atoms. It was this critical ability that enabled the subsequent experimental program.

Davis exposed the 3900-liter detector to higher antineutrino flux at the Savannah River reactor and again saw no signal above the nucleon-induced background ⁽¹⁰⁾. The upper experimental antineutrino-interaction limit was ~20% of the limit that would be expected if neutrinos and antineutrinos were the same. This result was an early indication that neutrinos and antineutrinos were not identical. Unfortunately, Davis did not publish these results, but only presented them in conference-report format at the 1958 Washington meeting of the American Physical Society ⁽¹¹⁾.

Before 1958, it did not appear likely that a reasonably sized Cl–Ar detector could detect neutrinos from the Sun. It was assumed that the dominant solar neutrino signal came from $p+p \Rightarrow D + e^+ + \nu_e$ with $E(\nu_e) < 0.42$ MeV and that the signal was not detectable because the energy threshold for the $^{37}\text{Cl} \Rightarrow ^{37}\text{Ar}$ transition was 0.81 MeV. Only the neutrinos from the CNO cycle were above the transition threshold and were therefore detectable. However, at the core temperature of the Sun, 15.6×10^6 K, the CNO cycle only contributes a few percent to the solar fusion rate. In his 1955 paper ⁽⁹⁾, Davis used the signal rate in the buried 3900-liter detector to place a limit on the solar neutrino flux from the CNO cycle at 1600 times the flux expected if all the solar energy were generated by CNO. The implication was that if we were to adjust for the diminished role of CNO, we would require a detector of 4×10^8 liters to detect those neutrinos.

Beginning the search for neutrinos from the sun

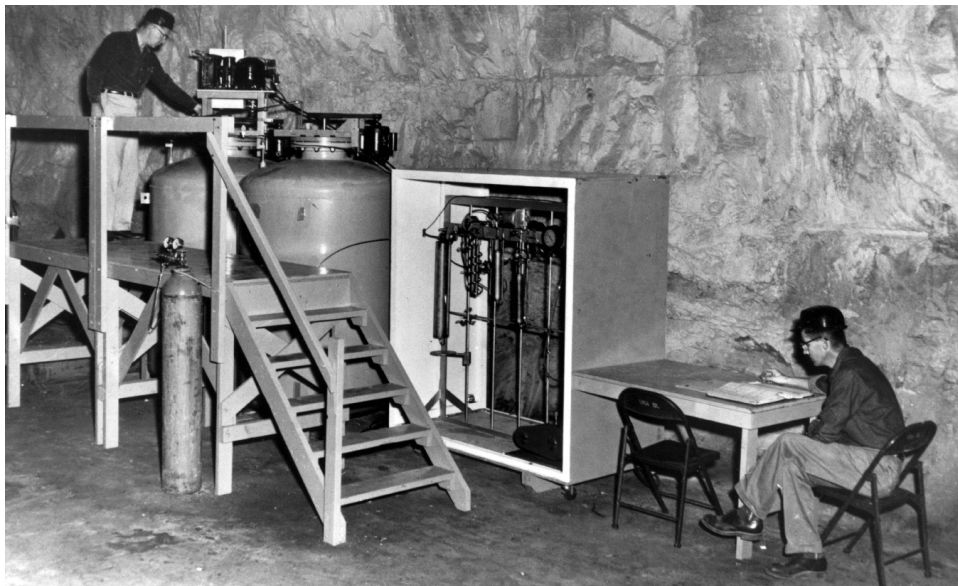
In 1958, Holmgren & Johnston ⁽¹²⁾ reported their measurements of the cross section for ${}^3\text{He} (\alpha, \gamma) {}^7\text{Be}$. Their measured cross section was $\sim 10^3$ times larger than had previously been assumed and thus predicted a significant rate of ${}^7\text{Be}$ production in the solar core. This measurement represented the critical breakthrough, as it provided a significant flux of neutrinos above the ${}^{37}\text{Cl} \Rightarrow {}^{37}\text{Ar}$ transition threshold. From the ratio of the cross sections of ${}^3\text{He} + {}^4\text{He} \Rightarrow {}^7\text{Be} + \gamma$ and ${}^3\text{He} + {}^3\text{He} \Rightarrow {}^4\text{He} + 2{}^1\text{H}$, and the relative concentrations of ${}^4\text{He}$ and ${}^3\text{He}$ in the solar core, it was determined that 15% of the ${}^3\text{He}$ resulted in the production of ${}^7\text{Be}$. Almost all of these transitions ended with $e^- + {}^7\text{Be} \Rightarrow {}^7\text{Li} + \nu$, where $E(\nu) = 0.86$ MeV. Approximately 10^{-4} of the ${}^7\text{Be}$ interacted with a proton and resulted in ${}^8\text{B}$, $p + {}^7\text{Be} \Rightarrow {}^8\text{B} + \gamma$. The ${}^8\text{B}$ then beta-decayed: ${}^8\text{B} \Rightarrow {}^8\text{Be} + e^- + \nu_e$. The energy spectrum of these neutrinos extended to 13 MeV.

As soon as Holmgren & Johnston ⁽¹²⁾ reported these measurements at the 1958 New York meeting of the American Physical Society, both William Fowler ⁽¹³⁾ and Alan Cameron ⁽¹⁴⁾ wrote letters to Ray Davis suggesting that he begin a search for solar neutrinos with his chlorine-based neutrino detector. There was great hope that the cross section for $p + {}^7\text{Be}$ to form ${}^8\text{B}$ would be large, that most of the ${}^7\text{Be}$ would be involved in this reaction, and that the 3900-liter detector would detect several solar neutrino interactions per day from the ${}^8\text{B}$ decay.

Because the expected signal was measured in counts per day rather than counts per minute, it was necessary to greatly reduce the cosmic ray background in the solar neutrino detector by placing the detector deep underground. At that time, Davis had two measurements of the ${}^{37}\text{Ar}$ -production rate by cosmic rays, one from the 3900-liter detector buried at a depth of 9.7 meters water equivalent (m.w.e.) at Brookhaven and another from an 11,700-liter detector located at a depth of 25 m.w.e. at the Savannah River facility. Both detectors yielded a cosmic ray-induced ${}^{37}\text{Ar}$ -production rate of ~ 70 counts per day per 3900 liters CCl_4 . Clearly, a much deeper location was required.

The PPG Corporation used a mine in Barberton, Ohio, to supply limestone for its glass production. At the depth of this mine, 1800 m.w.e. (2300 ft), the cosmic ray flux was ~ 2000 times smaller than in the previous shallow detector locations. Barberton Mine was beautiful and spacious, with very wide and deep chambers measuring 10 m high and with easy access to the surface. Davis, together with a Brookhaven technician, John Galvin, assembled a detector consisting of two 1950-liter containers, each of which contained an internal stirrer and a means for bubbling helium through the detection fluid. Instead of CCl_4 , they filled the detector with C_2Cl_4 , a less toxic chlorine-containing fluid.

The Barberton experiment was extremely simple. The setup consisted of the two tanks, a gas-handling apparatus, a small table, and two chairs—one for Davis and the other for Galvin.



The Barberton Mine detector. This 1963 picture shows the two 1950-liter perchloroethylene-filled vessels and the argon-extraction system in the housing to the right of those vessels. Ray Davis is on the platform used to access the vessel controls, and John Galvin is seated at the table, recording data in the notebook. Detector components that are not shown include the liquid nitrogen--containing dewars that surrounded the various traps in the argon-extraction system.

For the Barberton experiment, Davis augmented the ^{37}Ar -extraction and -purification system he had used in previous experiments with the addition of a carrier gas, ^{36}Ar , to measure the extraction and detection efficiency of the ^{37}Ar signal. Specifically, a small amount (0.1 cm³) of ^{36}Ar was added to the detector before the exposure. At the end of the exposure, the two argon isotopes, the ^{36}Ar carrier and the ^{37}Ar produced during the exposure, were extracted together and inserted into the counter. At the end of the counting period, the amount of ^{36}Ar in the counter-filling gas was measured. For each of the runs, the amount of ^{36}Ar recovered from the counter was equal to or greater than

95% of the amount inserted originally. The only assumption in this procedure was that the two argon isotopes were extracted with equal efficiency. ^{36}Ar is an ideal choice for a carrier gas because it is a very minor (1%) constituent of natural argon, which is mainly ^{40}Ar . Any in-leakage of argon from the atmosphere would be predominantly ^{40}Ar , and thus it would be easily distinguishable from the ^{36}Ar carrier gas extracted from the detector.

The earlier counting system, which used Geiger–Müller counters, had a background rate of 0.15–0.25 counts per minute, or ~ 17 per day, far too large for the Barberton detector signal. Davis developed a miniature proportional counter, 0.3 cm in diameter and 1.2 cm in length, that had approximately 4% of the volume of the previous counters. These proportional counters produced a pulse whose amplitude was proportional to the energy deposited in the counter. In this case, the ^{37}Ar decayed by orbital electron capture and emitted a 2.8-keV X-ray. The reduced detector mass and volume, together with the ability to select only pulses of the appropriate energy, drastically reduced the counter background rate. In addition, Davis surrounded the counter with a NaI-crystal anticoincidence system to eliminate Compton-scattered electrons. The result was an impressive 100-fold reduction in the background counting rate to 0.17 per day.

Unfortunately, there was no difference in counting rate between the proportional counters filled with normal argon and those filled with argon extracted from the Barberton experiment. All Davis could do was set an upper limit on the solar neutrino flux. By assuming that the entire counting rate of 0.17 per day was due to solar neutrinos, Davis set an upper limit on the product of neutrino flux multiplied by the neutrino cross section: $\phi\sigma < 3 \times 10^{-34}$ interactions s^{-1} , or 300 solar neutrino units (SNU, where 1 SNU = 10^{-36} interactions s^{-1})⁽¹⁵⁾.

While the Barberton experiment was under way, Ralph Kavanagh⁽¹⁶⁾ measured the cross section for $p + {}^7\text{Be} \Rightarrow {}^8\text{B} + \gamma$ and found it to be quite small. This rather discouraging result meant that Davis would need a much larger detector and probably a considerably deeper location for his experiment. In 1962, at the instigation of William Fowler, the California Institute of Technology (Caltech) Kellogg Laboratory hired a new postdoctoral fellow, John Bahcall, to work on the calculation of the solar neutrino capture rate of ^{37}Cl . Bahcall began a systematic evaluation of all the parameters that were involved, and by 1964 he had developed a detailed calculation of the capture rate. In January of that year, Bahcall published a paper⁽¹⁷⁾ adjacent to Davis's in *Physical Review Letters* in

which he predicted the rate of interaction between neutrinos from the Sun and ^{37}Cl as $\phi\sigma = (4 \pm 2) \times 10^{-35}$ interactions s^{-1} (40 ± 20 SNU) ⁽¹⁷⁾. Later that year, Bahcall published a more precise calculation of $\phi\sigma = (3.6 \pm 2) \times 10^{-35}$ interactions s^{-1} (36 ± 20 SNU) ⁽¹⁸⁾. Interestingly, most of this rate is due to a single transition from the ground state of ^{37}Cl to the 5.1-MeV excited state of ^{37}Ar . This superallowed transition was suggested by Ben Mottelson during a seminar Bahcall gave at the Niels Bohr Institute of Copenhagen in 1963.

The predicted interaction rate was a factor of ten smaller than the extreme lower limit that Davis set with the Barbarton detector. Clearly, a detector that was significantly larger than ten times the size of the detector at Barbarton was required. Davis pursued a 100-times-larger detector that could accommodate 390,000 liters. He found it difficult to convince both the funding agencies and the scientific community of the merit of constructing such a detector. The expected production, using the Bahcall calculation, was ~ 10 atoms of ^{37}Ar per day, or a few hundred such atoms in the detector after an exposure of several ^{37}Ar half-lives. It was hard to imagine that one could reliably extract a few hundred atoms from a detector containing $\sim 10^{31}$ atoms. Even if the extraction and counting could be done in a reliable and reproducible way, how would one distinguish ^{37}Ar atoms produced by solar neutrinos from those produced by local background processes or by cosmic rays that penetrated the Earth? Finally, the allocation of scientific financial resources had to be considered. Should the necessary funds come from the field of astronomy, physics, or chemistry? After all, Davis was in an unusual position: He was a radiochemist using physics techniques to answer a fundamental question in astronomy.

Fortunately, Davis had some very persuasive colleagues: John Bahcall, who was now devoting most of his scientific effort to this project; Dick Dodson, chair of the Brookhaven Chemistry Department, who had 15 years previously told Davis to find an “interesting problem to work on”; and William Fowler, who saw this experiment as the natural next step in determining the chain of nuclear reactions that occur in the core of stars. In addition, neutrino detection had come a long way since Pauli’s “embarrassing” and “desperate” suggestion ⁽³⁾ of 1930, which he had thought could never be experimentally proven. Cowan & Reines ⁽¹⁹⁾ had detected antielectron neutrinos from the Savannah River reactor in 1955, and another neutrino species—the muon neutrino—had been detected at the AGS accelerator at Brookhaven in 1962 ⁽²⁰⁾. This was a perfect time to look for neutrinos from the sky, that is, to begin the field of neutrino astronomy.

The homestake 390,000-liter perchloroethylene detector

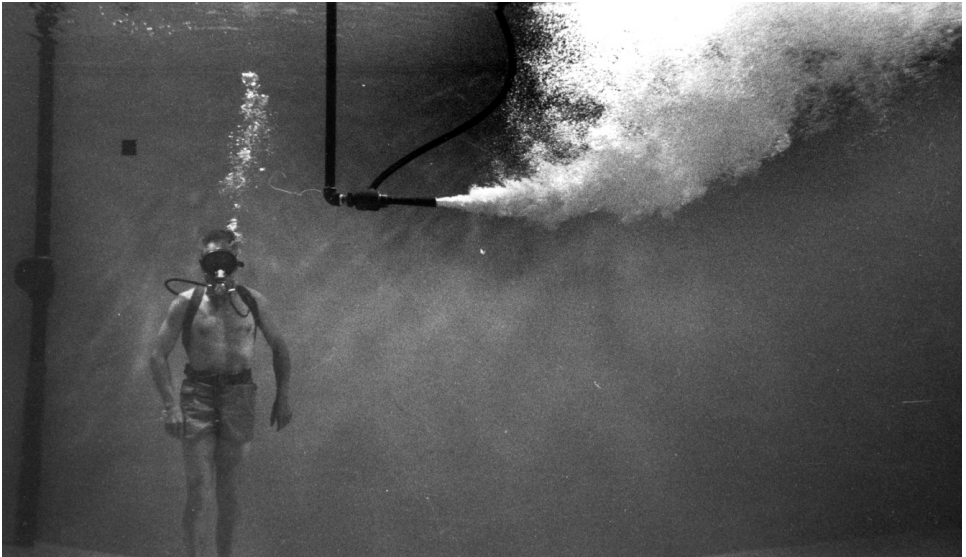
In his 1964 paper ⁽¹⁵⁾, Davis predicted a ³⁷Ar-production rate of 4–11 atoms per day, assuming Bahcall's most recent calculation of the interaction rate. At a depth of 4500 ft (4000 m.w.e.), the expected cosmic ray ³⁷Ar-production rate would be approximately a factor of 30 lower than the anticipated signal rate. Davis also described the need for a water shield to absorb neutrons from the surrounding rock and for necessary limits on thorium and uranium in the detector fluid. Not mentioned was the need for similarly low radioactivity in the construction materials of the detector. Davis even raised the issue of how to distinguish neutrinos of solar origin from other astronomical sources.

Finally, Davis needed a suitable underground location. There were not many mines in the United States that were 4500 or more feet deep and whose rock was sufficiently stable to permit the excavation of a large detector room. Two sites were seriously considered: the Homestake Mine in Lead, South Dakota, and the Sunshine Mine in Kellogg, Idaho. At first Homestake was not interested, so Davis turned his attention to the Sunshine Mine. Eventually, however, Homestake received some excellent scientific guidance from a member of its board of directors, and the company encouraged Davis to use its mine. That was a fortunate turn of events: Several years later, the Sunshine Mine had a serious underground fire.

Excavation of the chamber for the detector began early in 1965. The location chosen by the Mine was as far away from the mining activities as possible, but still relatively close to the main hoist to the surface, the Yates shaft. After the excavation was complete, Davis performed a careful neutron survey of the chamber with a calcium nitrate-filled detector, using the reaction $n + {}^{40}\text{Ca} \Rightarrow {}^{37}\text{Ar} + {}^4\text{He}$. Although the average neutron flux in the chamber was reasonably low, there was a "hot" region that resulted from an intrusion of rhyolite rock into one part of a side wall. Davis also carefully measured the radioactivity of the steel sections used to assemble the 6-m-diameter by 14.6-m-long steel tank, and he measured samples of the perchloroethylene detector fill at the chemical plant where it was being prepared.

In earlier detectors, helium from a gas cylinder was passed through the detector fluid, followed by a liquid nitrogen-cooled charcoal trap to remove the argon. The helium was then discarded. For the new detector, using this procedure would have involved the discharge of over half a million liters of helium per argon extraction. To avoid such excessive waste, Davis devised a new approach involving the recirculation of the flushing helium in a closed cycle. No new helium was introduced, and virtually no helium was lost.

This new system required pumping the perchloroethylene through a set of 40 eductors, Bernoulli-effect tubes that sucked in helium and mixed it with the perchloroethylene. Two special pumps whose rotors were immersed in perchloroethylene were constructed by Chempump. Davis tested the eductor system in the Brookhaven Lab swimming pool to ensure that the circular flow created by the eductors mixed the fluid throughout the tank. A reentrant tube that entered the tank from above provided a means of putting a neutron source into the center of the tank and thus could be used to calibrate the extraction efficiency of the entire system by generating a known number of ^{37}Ar atoms.



Testing the eductors for the Homestake Mine detector's argon-extraction system in Brookhaven National Laboratory's swimming pool. This picture shows Ray Davis in SCUBA diving gear checking the output of one of the eductors used to mix the helium with the perchloroethylene. The fluid is pumped through the left-hand vertical pipe. The fluid then passes through a narrowed section of pipe. The reduced pressure in that section then pulls in the flushing helium gas, which is connected via a winding hose. The Homestake Mine detector contained 40 such eductors, 38 of which recirculated helium within the detector and 2 of which brought in helium from the argon-extraction system.

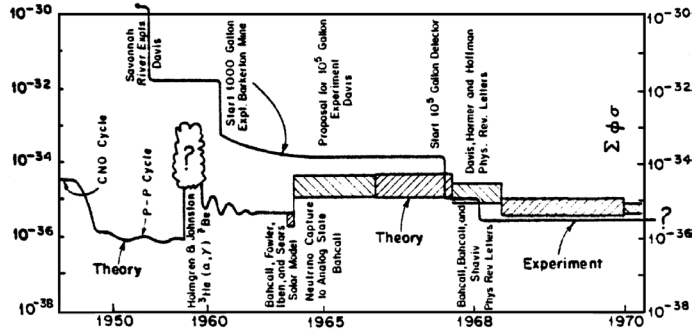
The solar neutrino problem: the observed signal is too small

In 1968 Davis published the results of the first two runs with the Homestake detector (21). In both runs, the counting rate obtained with the Homestake sample was comparable to the background counting rate. There was no signal above background, or $\phi\sigma \leq 3 \times 10^{-35}$ interactions s⁻¹ (3 SNU). In the meantime, using improved nuclear-interaction



The Homestake Mine detector in 1966. Ray Davis is standing on the walkway at the top of the detector, and John Galvin is at the bottom of the ladder. The chamber was subsequently filled with water to just below the top walkway so that the entire detector was submerged. This water fill provided a shield against neutrons from the excavation walls.

parameters, Bahcall (22) recalculated the expected rate as 20 SNU, so that the observed rate was less than one-seventh of the new predicted rate. Was there a problem with extraction efficiency? In the first two runs, Davis recovered 94% and 95%, respectively, of the originally inserted ³⁶Ar carrier gas. Was there a problem with the predicted cross sections and fluxes? Bahcall carefully reviewed the calculations and found no error (see figure below for a typical comparison of predicted signal versus observed signal). Was either the Sun or the neutrinos more mysterious than they had thought?



A viewgraph from a talk given by Ray Davis in 1971. Shown are the timelines of both the observed (experimental) and the predicted (theoretical) solar neutrino signal from a chlorine-based detector. The vertical scale is $\phi\sigma$ (solar neutrino flux \times ³⁷Cl cross section), where $10^{-36} = 1$ SNU. The papers named in the figure correspond to References 9–12, 15, 17, 18, 21, and 22.

Davis now faced three challenges. First, because the upper limit on the signal was more than an order of magnitude smaller than the predicted signal, it would have been desirable to increase the detector mass by an order of magnitude, but that was impossible both financially and logistically. In the original experimental plan, the expected cosmic ray–induced ^{37}Ar production was a factor of 30 smaller than the solar signal. Now, the cosmic ray background could be similar in magnitude to the solar signal. Because it was impossible to move the detector to a deeper location where the cosmic ray flux was smaller, Davis embarked on a campaign to accurately determine the cosmic ray background, which involved building several smaller, portable C_2Cl_4 detectors and exposing them at various shallower levels in the mine. This campaign took a number of years and was effectively limited to depths of 1800 ft or less because the cosmic ray signal was not detectable in the smaller tanks at deeper locations. The extrapolation to the 4850-ft depth of the rate at which cosmic rays produced ^{37}Ar was carried out with the help of Arnold Wolfendale, E.C.M. Young⁽²³⁾, and George Cassidy⁽²⁴⁾. Unfortunately, the uncertainty in the cosmic ray–induced rate remained the largest error in the measurement and would have required a major and possibly unwarranted effort to improve.

The second challenge was to improve the counter event selectivity. Here serendipity played a role. In 1968, while Davis was visiting Caltech, he got into a conversation with another physicist, Gordon Garmine, at the campus swimming pool. As Davis was describing the counter-selectivity situation, Garmine suggested that, in addition to his proportional counter energy measurement, he also measure the pulse rise time. The ^{37}Ar decay, which involves the capture of an orbital electron and the emission of Auger electrons, is a localized event and so should produce a fast-rising pulse on the counter center wire. The dominant counter background is due to Compton-scattered electrons that traverse the counter and deposit energy at a range of distances from the center wire. These result in a slow-rising pulse. When Davis returned to Brookhaven, he asked the Instrumentation Group to design and build pulse rise–discrimination circuitry. With this new component, the signal-to-background discrimination improved considerably so that the counter background rate in the energy–rise time region of interest was reduced to ~ 1 count per month.

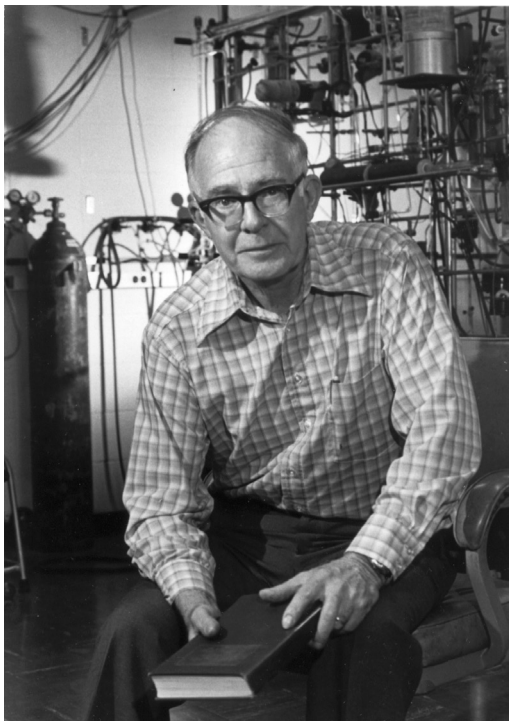
The third (and least significant) challenge was to convincingly demonstrate that the process of extracting argon atoms from the 390,000-liter tank and transporting those atoms to the proportional counter was highly efficient. Davis's original extraction-monitoring technique was to insert into the tank a small, known amount of ^{36}Ar and then measure the fraction of the isotope that was recovered after extraction and counting.

He now modified this technique by alternating ^{36}Ar and ^{38}Ar carrier gas in adjacent extractions. Specifically, he inserted ^{36}Ar into the tank for the first extraction, then repeated the process for the next extraction with ^{38}Ar . In each case, he measured the amounts of both ^{36}Ar and ^{38}Ar in the extraction. Thus, if the first extraction yielded 95% of the inserted ^{36}Ar , the next extraction should yield the remaining 5% of ^{36}Ar . This procedure ensured that no reservoir of carrier gas remained in the tank. Results obtained from the two alternate carriers agreed with the earlier conclusion that almost all of the argon in the tank was being extracted.

There was one other extraction-related concern: What if there were regions of the tank interior that the isotopic carrier never reached and that were also unaffected by the extraction process? Fortunately, Davis's meticulous record-keeping and measurements addressed this concern.

When the C_2Cl_4 was first brought to the mine, it contained dissolved argon and other gases. Davis performed a series of helium sweeps of the tank and measured the amount of gas extracted during each of these sweeps. The extracted gas volume decreased exponentially. If there had been a "dead" (i.e., inefficiently swept) region in the tank, this exponential decrease would have exhibited two slopes, one for the efficiently swept region and another for the dead region. Because the extraction plot showed only a single slope, the dead-region concept was ruled out.

When I first visited the experiment in the Homestake Mine in 1972, Davis was considering a complete test of the detector using a neutrino source placed in the center of the tank via the reentrant tube. The source he considered, ^{65}Zn , decays by orbital electron capture and produces 1.25-MeV monochromatic neutrinos. The idea was to produce this isotope via $n + ^{64}\text{Zn} \Rightarrow ^{65}\text{Zn} + \gamma$ at a reactor at Oak Ridge National Laboratory. After considerable effort, it became clear that production of a source of sufficient strength



Raymond Davis, Jr. in front of part of the argon gas-extraction system (taken in 1978).

was not feasible at that reactor. Instead, Davis employed a simpler mode of producing a known amount of ^{37}Ar in the tank. In this approach, a neutron source, PuBe, was introduced into the reentrant tube, and ^{37}Ar was produced by $^{35}\text{Cl}(n,p)^{35}\text{S}$ followed by $^{37}\text{Cl}(p,n)^{37}\text{Ar}$. The amount of ^{37}Ar extracted after this exposure was consistent with that predicted by the source strength, the interaction cross section, and the exposure time.

Several unorthodox explanations for the reduced signal were proposed. One suggestion was that because of the small momentum transfer involved in the $^{37}\text{Cl}(\nu, e^-)^{37}\text{Ar}$ reaction, an ^{37}Ar ion remained trapped in the original molecule, that is, a $\text{C}_2\text{Cl}_3\text{Ar}$ molecule. Davis even devised a test for this hypothesis: He used the Brookhaven reactor to make C_2Cl_4 in which one of the chlorine atoms was ^{36}Cl . ^{36}Cl beta decays to ^{36}Ar , with a momentum transfer comparable to that involved in the solar neutrino interaction in $^{37}\text{Cl}(\nu, e^-)^{37}\text{Ar}$. The rate of ^{36}Ar recovered in this test detector was consistent with the number of ^{36}Cl ions produced by the reactor exposure and with the lifetime of ^{36}Cl . No “low transfer-momentum” trapping of argon atoms occurred.

While Davis was methodically reviewing and improving all the experimental aspects of neutrino detection, theorists were endeavoring to review and improve their understanding of the fusion cycles in the solar core and the predictions of the solar neutrino flux. Bahcall carefully and systematically reviewed all the nuclear physics input information required for both the solar fusion reactions and the neutrino interaction cross section for ^{37}Cl (25). As these various parameters changed, so did the prediction rate. However, the prediction rate tended to hover around 8 SNU, approximately one-quarter of the original prediction and approximately three times the observed rate in the Homestake Mine detector. By 1975, enough solar neutrino-produced ^{37}Ar had been accumulated that it was possible to observe the ^{37}Ar lifetime from the total of all detected events. This development, although not unexpected, helped to confirm the identification of the signal and improved the precision of the signal.

A completely different explanation of the low solar neutrino signal was developed by Pontecorvo. In 1958, just after Davis presented the Savannah River experimental results, Pontecorvo suggested that there may be oscillations between neutrinos and antineutrinos analogous to the oscillations in the K^0 system (26, 27). This suggestion was made four years before the muon neutrino was discovered, so the only possible neutrino oscillation was between electron neutrinos and electron antineutrinos. In 1977, after the muon neutrino was discovered and after Davis had presented a reduced but positive solar neutrino signal, Pontecorvo revised his suggestion to oscillation between electron

neutrinos and muon neutrinos. Pontecorvo was the first to identify the source of the reduced solar neutrino signal, but it would be another decade before the world began to embrace the concept of neutrino flavor oscillation.

Gallium solar neutrino detectors

One of Davis's lingering suspicions was that ^{37}Cl detection rate's strong dependence on (a) a single transition, the superallowed transition from the ground state of ^{37}Cl to the 5.1-MeV excited state of ^{37}Ar , and (b) the relative production rate of ^8B in the Sun could result in a misleading signal expectation. Davis, together with Keith Rowley, had begun developing a detector using the reaction $^7\text{Li}(\nu e, e^-)^7\text{Be}$, the inverse of one of the solar fusion reactions. The threshold for this reaction is 0.86 MeV, so it is clearly sensitive to the neutrinos from ^8B as well as to that part of the ^7Be ($e^-, \nu e$) ^7Li that is above the terrestrial threshold. However, detection of ^7Be is difficult and was never developed well enough to employ this detector.

What Davis needed was a way to detect the neutrinos from the primary solar fusion reaction, $p+p \Rightarrow D+e^++\nu_e$. In 1965, Kuzmin ⁽²⁸⁾ pointed out that $^{71}\text{Ga}(\nu e, e^-)^{71}\text{Ge}$ had a threshold of 0.223 MeV, considerably below the 0.42-MeV end point of the $p+p$ fusion reaction. Because the solar thermal output depends critically on the $p+p$ reaction rate, the prediction of the $p-p$ fusion rate was very reliable and robust. However, in 1965, gallium was a rare material and was available only in limited quantities. Nonetheless, in 1974 we started working on a prototype gallium solar neutrino detector. We began by borrowing 50 kg of gallium from a U.S. Department of Energy facility, and that summer we formed two construction teams. Davis and Bill Frati, from the University of Pennsylvania (Penn), built a detector that used GaCl_3 dissolved in HCl acid, and John Evans and I constructed a metallic gallium detector. The goal for each system was to extract the germanium, then convert it into a gas (GeH_4) that could be counted in a proportional counter. Both techniques worked, and by the end of the summer of 1974, we had two working models at the several-kilogram scale.

The technical challenge we faced was to convert the benchtop gallium experiment into a 30–50-ton detector and then to fund this effort. The funding hurdle was significant because gallium then cost \sim \$500,000 per ton, or \$20–25 million for a full-size detector. Of course, the gallium would not be consumed during the course of the experiment, so the material cost could be recovered at the end of the experiment. Also, the chlorine experiment had always operated with a very small group: Davis and an additional two or three scientists. However, given the size of this new detector and the costs involved, a

considerably larger group was formed, and funding for a 1.5-ton prototype detector was obtained. By mid-1977, this scaled-up detector was operating, and it was time to fund the full-scale solar detector. To that end, we published a paper describing the results of the prototype work and the motivation for the full-size detector ⁽²⁹⁾ and held a workshop on new solar neutrino experiments Brookhaven Lab in January, 1978 ⁽³⁰⁾.

Our attempt to fund a U.S.-based gallium detector was unsuccessful, even though a very impressive review panel, chaired by Glenn Seaborg, recommended that it proceed. As part of the funding attempt, an international collaboration involving a strong group at Heidelberg was formed. The plan was for one-quarter of the gallium to be purchased with German funds and the rest with U.S. funds. Because Germany appropriated the necessary funds and the United States did not, the Heidelberg group then formed a mainly European collaboration, GALLEX, which then built a GaCl₃-based detector at Gran Sasso. Part of the Brookhaven-based collaboration eventually joined GALLEX. Meanwhile, the Soviet Union had produced 60 tons of metallic gallium. A group based at the Institute for Nuclear Research in Moscow arranged to borrow this material and built a gallium metal-based detector in an underground laboratory at Baksan in the Caucasus. In 1984, the Soviet collaboration invited us to join their experiment. Thus, both of the techniques developed at Brookhaven in 1974 eventually found use.

Moving from Brookhaven to Penn

In 1984, Davis turned 70, the mandatory retirement age at Brookhaven Laboratory. In his honor, we held that year's International Solar Neutrino Conference ⁽³¹⁾ in Lead, South Dakota, the site of the Homestake Mine. Such a large conference was an unusual event for this small mining town. The meeting was to be held in the Homestake Opera House, a beautiful building dating from the early 1900s. Unfortunately, two months before the conference, there was a fire in the Opera House. Luckily, the local high school was made available and we held the conference there.

The conference was staffed by the wives and some of the children of the chlorine group scientists. The enthusiasm of the local community knew no bounds. One evening, the conference participants attended a local theater production about a shooting that took place a century ago, *The Trial of Jack McCall*. Part of the show consisted of forming a jury from the audience. The jury that evening included Willy Fowler, Fred Reines, and Ray Davis. Jack McCall never had a chance and was convicted. The Homestake Mining Company also agreed to conduct tours of our underground laboratory for all the

conference participants. Normally, Davis hosted an underground meal for visiting scientists, but that proved impossible given the number of visitors.

The conference had an important outcome: The local community and the State of South Dakota became aware of the significance of Davis's solar neutrino detector in the Homestake Mine and of the worldwide scientific interest in neutrinos. This awareness eventually became the basis of South Dakota's successful offer to host the forthcoming Deep Underground Science and Engineering Laboratory (DUSEL) at the Homestake Mine.

However, in 1984 several problems developed. The first was the failure of the Homestake detector's two perchloroethylene-circulating pumps. These pumps were specially designed to pump perchloroethylene through the tank and extraction system without allowing the chemical to have any contact with the air. To do so, the rotor of the pump had to be completely immersed in the perchloroethylene, with a thin metal cylinder separating the rotor from the stator windings of the pump. The stator windings were immersed in oil in order to cool them. Each of these pumps operated for ~ 25 h per extraction, or only ~ 1500 – 2000 h throughout the 18 years between 1966 and 1984. However, the stator insulation had been exposed to the cooling oil during these 18 years, and the oil had penetrated through pinholes in the insulation. Thus, one pump failed in early 1984 and the other later the same year.

Second, the U.S. Department of Energy decided that because Davis was about to retire, the experiment would be terminated. Fortunately, Penn appointed Davis to the position of research professor and provided funds for a replacement perchloroethylene-circulation pump. Even better, after some consideration, the National Science Foundation agreed to take over support of the experiment. Thus, by 1986 solar neutrino data was again being taken by the chlorine detector.

The mid-1980s saw a number of exciting and critical developments in neutrino astronomy. On the experimental front, Kamiokande, a water Cherenkov solar neutrino detector, reported its first results: a detected 8B neutrino rate that was slightly less than half the predicted rate⁽³²⁾. Because both the detection technology and the energy threshold of Kamiokande were different from those of the Homestake Mine detector, this new observation definitely confirmed either that the predicted solar neutrino flux was incorrect or that there was another mechanism operating to reduce the size of the detected signal. The difference between the Kamiokande and Homestake detection rates also generated much interest. Was this difference a problem with the experiments, or was it an indication of as-yet-unknown fundamental physics?

Numerous theorists examined the predicted fusion reaction chain in the Sun to determine by how much they could reduce the predicted flux. The exercise was impressive and, at the extreme, managed to reduce the predicted flux by almost a factor of two. That reduction was sufficient to provide an overlap with the early Kamiokande results, but not with the Homestake detector results. Again, whether or not systematic effects or other corrections needed to be applied remained a mystery. This uncertainty was to continue for another decade.

Neutrino flavor oscillations: The MSW effect

Another approach, which had initially been of limited interest, involved reexamining the neutrino flavor oscillations proposed by Pontecorvo. In 1978, Lincoln Wolfenstein (33) attempted to determine (a) whether the effective mass of electron neutrinos passing through matter differed from that of muon neutrinos and (b) whether this mass difference could lead to oscillations from one flavor to another. His initial study included only two neutrinos, as detection of the third neutrino, the tau neutrino, was still under way. The two concepts, Pontecorvo's neutrino flavor oscillation in vacuum and Wolfenstein's matter oscillation, were combined into a single theory by Mikheyev & Smirnov⁽³⁴⁾ in 1985. In its simplest version, The Mikheyev–Smirnov–Wolfenstein (MSW) matter oscillation theory predicted that solar neutrino emission consisting entirely of ν_e in the solar core converted into one-third ν_e , one-third ν_μ , and one-third ν_τ by the time it reached the Earth. Because only ν_e can convert ^{37}Cl into ^{37}Ar , the chlorine detector should observe one-third of the neutrino flux emitted in the Sun, exactly what was observed. However, in the water Cherenkov detector, the signal arose from neutrino-electron elastic scattering. In addition to the normal charged-current elastic scattering of ν_e , there was also the neutral-current scattering of ν_μ and ν_τ from electrons, with a cross section one-sixth of the charged-current cross section. Therefore, the signal in Kamiokande should measure $\frac{1}{3} + \frac{1}{6}\left(\frac{2}{3}\right)$, or 0.44, of the predicted signal. Finally, the MSW theory provided a physical explanation of both the reduced observed flux and the apparent difference between the signal sizes from the two operating detectors. Of course, there were additional parameters to determine and other factors to reconcile.

Davis and the chlorine program were now in the third phase of the experiment, the need to obtain as precise a result as possible. Upon Davis's move to Penn, his Brookhaven group split up. Bruce Cleveland, who had joined Davis in 1976, moved to Los Alamos, and Keith Rowley, who had worked on both the lithium and the gallium detectors, stayed at Brookhaven and eventually joined the GALLEX experiment at Gran Sasso. Fortunately, there were excellent graduate students at Penn. Two outstanding graduate

students, James Distel and Paul Wildenhain; a Penn research scientist, C.K. Lee; and several undergraduate students joined the experiment. In addition, two of Davis's good friends, Jack Ullman from Lehman College and Ed Fireman from the Harvard-Smithsonian Astrophysical Observatory, also participated in the experiment.

The first apparatus we updated was the counter-readout system. In the mid-1960s, when this system had been constructed, the easiest way to record the data was with paper tape. When the counting system was moved to the mine, that system was still in use. We now installed a computer-based readout system that allowed telephone data transmission to Penn. Many more data could now be taken and transmitted, especially for the periodic counter calibrations. Wildenhain developed a rigorous set of event-selection criteria, then tested them against the criteria employed for the earlier data sets. The main effect was a more precise and reproducible extrapolation of backgrounds from the slow-rising pulse region, dominated by Compton-scattered electrons, into the fast-rising region where the ^{37}Ar -decay events lay. The systematic error for this background correction was significantly reduced.

Next, Jim Distel remeasured the internal efficiency of each of the proportional counters. With the help of Wick Haxton and Eric Adelberger, we obtained a sample of ^{127}Xe from the University of Washington. Whereas ^{37}Ar generally decays to the ground state of ^{37}Cl , ^{127}Xe never decays to the ground state of ^{127}I . Instead, approximately half of ^{127}Xe 's decays go to the 375-keV excited state of ^{127}I and then to the ground state through the emission of two gamma rays. Thus, we had a threefold coincidence in the decay—a very distinctive signature. We placed each counter into a well inside a split NaI detector, that is, a detector in which the two half cylinders were light-isolated from each other and read out separately. We selected events in which there were coincident gamma rays in each of the two NaI crystals and measured the probability of any pulse appearing in the proportional counter in coincidence with the two gamma rays, the proportional counter efficiency. Next, we looked at the height and shape distributions of these proportional counter pulses to determine the fraction that would meet our signal to background discrimination procedure. From the latter, we determined the pulse-height distribution of events in the fringe field of the proportional counters. Using these data, we designed and tested a new set of counters whose fringe region contained guard rings. These counters had a larger ^{37}Ar -detection efficiency, which was equivalent to having a larger solar neutrino detector, but at a much lower cost. Finally, we repeated the calibration process several times for a number of counters, then used the scatter of efficiencies from the repetitions to determine the systematic error, $\sim 0.5\%$. Of course, we were constrained by the ^{127}Xe half-life of 36 days.

When the counters were taken underground again, we noticed that a number of them had higher backgrounds. Upon comparing the before and after background pulse-height distributions, we realized that while the detectors were at the surface of the earth, cosmic rays had induced a small ^{55}Fe contamination. The cathode cylinder was made of highly purified iron, which had been activated by cosmic rays. We reviewed the background data of the counters from the time each was first brought into the mine a decade earlier and found the same effect in the earlier data. Although the effect was small ($\sim 2\%$), we made the appropriate correction. Finally, we cross-calibrated the gas-volumetric measure used for the insertion of carrier gas with that used for the extracted sample.

As a result of these careful calibrations, we reduced the systematic error associated with the system to less than 5%. During this period, we also maintained a continuous set of ^{37}Ar extractions from the detector so that by the late 1990s the statistical error was also less than 5%. Davis had designed his detector so well that even after the signal was reduced by an order of magnitude it was still possible to obtain a very precise measure of the solar neutrino flux. The dominant error, which we could not reduce, was due to cosmic rays. Although the signal shrank by an order of magnitude, the cosmic ray background did not.

In the late 1990s we began to continuously extract and collect the argon atoms in two 12-h traps, one for those produced during the day, when the path from the Sun passes through a small thickness of the Earth, and another for those produced at night, when the path from the Sun passes through a thickness of the Earth nearly equal to its diameter. The manually filled, liquid nitrogen-cooled charcoal trap was replaced by a cryorefrigerator, which required considerable reconstruction of the extraction and gas-collection systems. The experiment was ready for operation by mid-2000. Unfortunately, on September 11, 2000, the Homestake Mining Company announced that because of the low price of gold, it would cease mining operations at the Homestake Mine.

Epilogue

One window closed: The experiment that began so dramatically in 1965 had ended. However, another window opened: There was now a natural location for a new and much-needed facility for the United States, DUSEL. Thus, chlorine-based solar neutrino detection died, and DUSEL was born.

Raymond Davis's great achievement in neutrino astronomy was recognized around the world. His first prize was the 1957 Boris Pregel Prize from the New York Academy of Sciences. In 1978, he received the Comstock Prize from the U.S. National Academy of Sciences, followed by the American Chemical Society Award for Nuclear Applications in Chemistry in 1979, the American Physical Society's Tom W. Bonner Prize in 1988, and the W.K.H. Panofsky Prize in 1992. He received honorary doctorates from Penn in 1990, Laurentian University in 1997, and the University of Chicago in 2000. The American Astronomical Society awarded Davis the Beatrice M. Tinsley Prize in 1995 and the George Ellery Hale Prize in 1996. Russia awarded him its first Bruno Pontecorvo Prize in 1999, and Israel awarded Davis the Wolf Prize in 2000. Davis was awarded the U. S. National Medal of Science in 2001 and the Franklin Medal in 2003. On December 10, 2002, Davis shared the Nobel Prize in Physics with Masatoshi Koshiha and Riccardo Giacconi.

Raymond Davis, Jr. died peacefully at his home in Blue Point, New York, on May 31, 2006.

In this historical review, I have tried to describe in parallel Raymond Davis's scientific activities and the growing understanding of neutrino emission from the solar core. These two tracks were closely coupled. Without the stimulation of Davis's experimental results and John Bahcall's continuing modeling efforts, it is unlikely that our understanding of neutrinos would be what it is today. What I have omitted to mention so far, however, is my personal relationship with Ray. I first met him at a neutrino workshop at Los Alamos in December 1970, and for the next year or so I periodically visited him at Brookhaven. During an American Physical Society meeting in spring, 1972, Ray suggested that I come out to Homestake. A week later I did, and I spent the next 30 years working with him. It was an extraordinary experience. Ray had an uncanny, intimate understanding of experimental apparatus and systems. He and the detector seemed almost to be one. He carried out measurements with great precision and care, and his notebooks were magnificently organized. Coffee breaks and lunches in the underground laboratory were occasions for discussion of critical scientific topics. Some of these discussions were

resolved quickly, others persisted for long periods or forever, but all were stimulating and engaging. Our students learned from his example how science and scientists functioned.

Davis was also a vigorous and athletic person. He spent Sundays at Homestake climbing to the top of the peaks in the Black Hills, exploring streams to see where they came from, swimming in the many lakes, and, in the winter, walking out over their frozen surfaces. He was enthusiastic about every aspect of life and lived every moment to the fullest.

Above all, what was supremely impressive about Ray was his personality, his interest in science, his kindness to all, his pleasant demeanor, his tolerance of others, and his gentleness. He was more than a great scientist; he was a great person. Ray Davis is sorely missed.

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