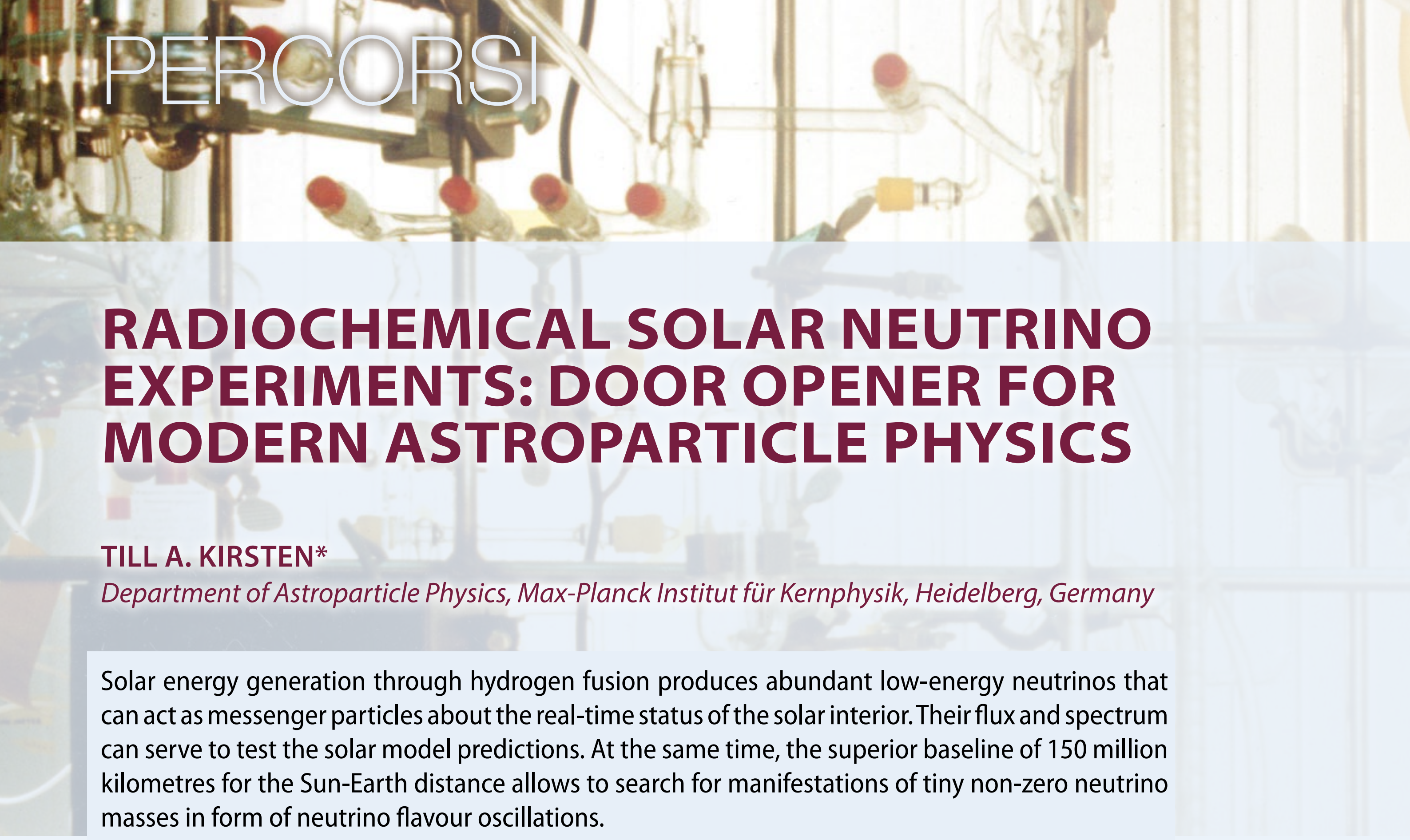

RADIOCHEMICAL SOLAR NEUTRINO EXPERIMENTS: DOOR OPENER FOR MODERN ASTROPARTICLE PHYSICS

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Solar energy generation through hydrogen fusion produces abundant low-energy neutrinos that can act as messenger particles about the real-time status of the solar interior. Their flux and spectrum can serve to test the solar model predictions. At the same time, the superior baseline of 150 million kilometres for the Sun-Earth distance allows to search for manifestations of tiny non-zero neutrino masses in form of neutrino flavour oscillations.

Solar neutrino detection is a formidable experimental challenge because of the extremely small interaction cross sections. This narrative describes the development of the hunt for solar neutrino detection that lasts now for more than 50 years, with emphasis on the first 35 years from 1962 to 1997. This path making period is marked by "radiochemical" experiments with very large Chlorine and Gallium detectors that were installed in deep underground laboratories for suppression of background reactions. The encouraging results from the radiochemical experiments intensified the later development of modern super-sized real-time detectors. Neutrino astrophysics is now a flourishing field, competitive and complementary with accelerator particle physics. Long forgotten, this was triggered by radiochemical experiments.

1 How it all started

Neutrinos are the most abundant elementary particles in the universe, even more abundant than photons. Notwithstanding, for long they remained mysterious because of their elusive properties: seemingly massless, without electric charge, and without any strong interaction with matter. To begin with, they were not even discovered but just postulated by Wolfgang Pauli in 1930 to account in a hypothetical way for the missing energy in the continuous spectra of radioactive nuclei that emit electrons (Beta-particles). Such beta decays are the major manifestation of the Weak Force, the least familiar of the four fundamental interactions that rule our world (the others being gravity, electromagnetism, and the strong nuclear force, that governs alpha radioactivity, fission and nuclear reactions). It is in these Beta processes that neutrinos come into being by virtue of weak interactions.

A hypothetically invented particle longs for experimental detection, but with no manifest interactions, this wish seemed to be an illusion. Following Enrico Fermi's theory of Beta decay, the cross section of a MeV neutrino beam with matter was estimated to be so small – $O(10^{-44}\text{cm}^2)$ – that it could penetrate the whole Earth or the whole Sun without any measurable absorption. However, in 1946 Bruno Pontecorvo (fig. 1) overcame the resignation of hopelessness for neutrino detection by suggesting that the signal rate in an attempted experiment could be greatly enhanced by not looking for the individual interactions but using a detection scheme that is based on inverse Beta decay in a huge detector where the small individual reaction cross section is (at least in part) compensated for by a very large number of target nuclei. Inverse

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Fig. 1 Bruno Pontecorvo and Till Kirsten at Selinunte Temple, 1989

Beta decay implies that a target nucleus with *Z* protons absorbs a neutrino and forms, with the emission of an electron, a product nucleus *Z*+1. In the same paper, Pontecorvo made a very practical suggestion for the case of taking ^{37}Cl as the target: $^{37}\mathrm{Cl}\ (\nu, e^-)\ ^{37}\mathrm{Ar}$.

The historical quote in the famous 1946 "Chalk River Report" [1] reads:

"The experiment with Chlorine, for example, would consist in irradiating with neutrinos a large volume of Chlorine or Carbon Tetra Chloride, for a time of the order of one month, and extracting the radioactive Ar37 from such volume by boiling. The radioactive argon would be introduced inside a small counter… "

^{37}Ar has a half life of 34 days. If one follows its decay for a few months in a counter with very low background ("Low-Level-Counter"), the neutrino-induced production rate can be determined.

This defined once and forever the principles of what became later known as the "Radiochemical Method" of rare event detection. The accumulation of interaction products during a rather long collection time lowers the detection limit dramatically. The prize to pay for this is the loss of the kinematic details of the individual events. It is then mandatory to exclude disturbing backgrounds and side reactions that could mimic the rare reaction under study (*e.g.* neutrino capture). This crucial aspect is already considered in the Chalk River report.

Pontecorvo's intention was to experimentally assure the questionable existence and the fundamental properties of the neutrino, whether it exists at all, and what is its role in the overall scheme of elementary particles. Astrophysical objects appeared on the scene as potentially strong neutrino sources in experiments to investigate neutrino properties. It was well known since the works of Sir Arthur Eddington that stars (like our Sun) produce most of their energy by nuclear fusion reactions. If neutrinos existed at all, this would make stars very intense neutrino sources. Other than photons, neutrinos would freely escape the stellar core where they were produced, testimonial about the conditions at their origin – a real time look into the stellar centre!

In 1946, for the detection of neutrinos Pontecorvo considered primarily a nuclear reactor as neutrino source, but since the conception of lepton number conservation (the distinction between neutrino and antineutrino) was not established at the time, he also considered the possibility to use the Sun as a neutrino source for detection. However, he dismissed this possibility because of inappropriate flux and cross section estimates. Notwithstanding, the fascinating idea of looking directly into the solar core and to experimentally verify hydrogen fusion as the prime source of stellar energy initiated closer examinations by H.R. Crane from Caltech and – in particular detail – by Luis Walter Alvarez from Berkeley University [2]. Many questions remained open, both concerning theoretical principles and very uncertain production rate predictions. In spite of this, in 1958 Raymond Davis jr. at the Chemistry Department of Brookhaven National Laboratory (BNL) started the first experimental realizations of the ^{37}Cl-^{37}Ar detection scheme with a 1000

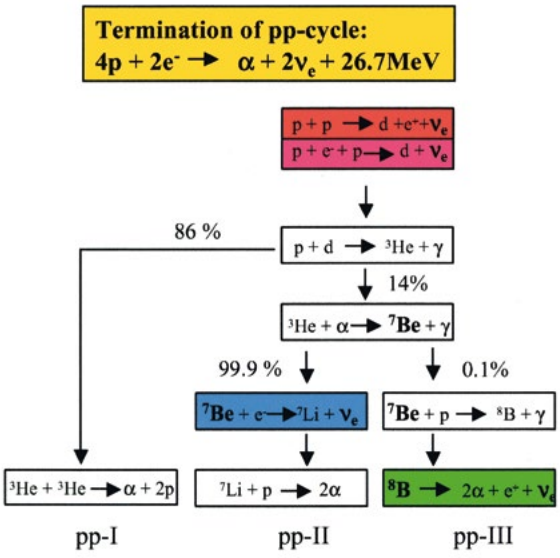

Fig. 2 Solar fusion reaction chains.

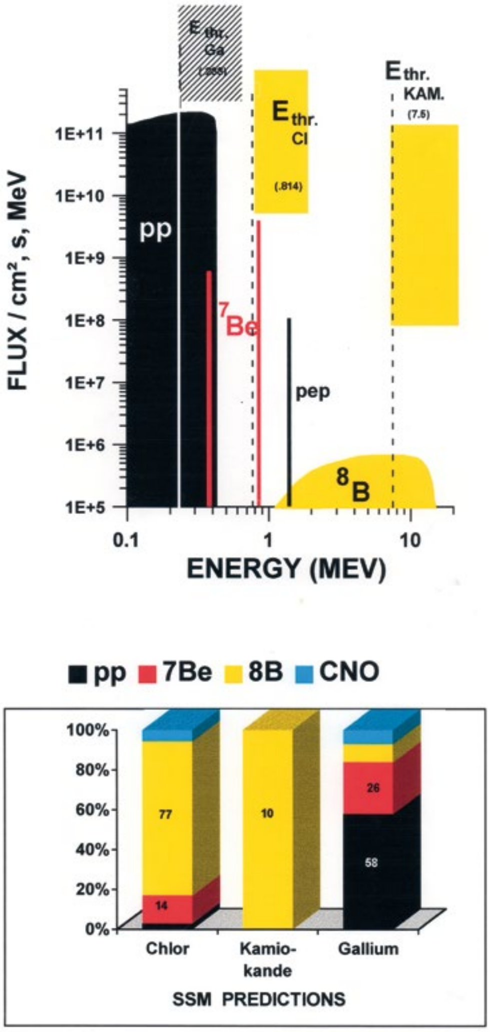

Fig. 3 Top: Solar neutrino spectrum expected from the Standard Solar Model (SSM). Bottom: Expected percentage participation of neutrino types to the expected signal without propagation effects in key solar neutrino experiments.

gallons perchlorethylen (C_2Cl_4) experiment that he exposed at the Savannah River power reactor plant, with discouraging results. But the experiment defined the prerequisites that needed to be addressed: working in deeply shielded underground laboratories, hyper purity for consequent background reduction and ultimate low level techniques.

2 Feasibility of a solar neutrino detector

Main sequence stars like our Sun produce neutrinos while they generate their energy through slow hydrogen burning over billions of years. A first estimate of the expected pp-neutrino flux comes from the well-known solar luminosity. In each fusion chain $4H \rightarrow {}^4He$ that is completed in the solar core, 26.73 MeV of energy and 2 neutrinos are generated. The core temperature T_c is 15.8 million K. On average, 0.59 MeV escape with neutrinos; 26.14 or 13.07 MeV per pp-neutrino make the Sun luminous. Dividing the solar luminosity by 13.07 MeV yields the total pp-neutrino flux. At the Earth, 150 million km away from the source, this flux is $6 \times 10^{10}\,\mathrm{cm}^{-2}\,\mathrm{s}^{-1}$ [3]. It depends only moderately on the central temperature ($\sim T_c^{-1}$).

In spite of this very high flux, for a long time a solar neutrino experiment seemed unrealistic because of the extremely small cross sections. In addition, the low energy of pp-neutrinos (maximum 420 keV) excluded altogether the option to detect them with a ${}^{37}Cl$-${}^{37}Ar$ detector since its energy threshold is 814 keV. There remained the vague possibility to settle for the higher energetic ${}^8B$ neutrinos from the rare PPIII side branch of the solar fusion reaction network (see fig. 2), but their flux is much lower, less well predicted and the contribution of this branch to the solar luminosity is negligible.

Against all odds, two scientists did not give up but rather devoted their further scientific carrier to this outrageous task, Ray Davis as experimentalist and John Bahcall as theoretical astrophysicist at the Princeton Institute for Advanced Studies.

First of all, a reliable prediction for the expected solar neutrino fluxes had to be deduced from the best available solar model (Standard Solar Model, SSM).

Various specific reaction chains produce a neutrino spectrum in the energy range of $O(10^{-1} - 10)$ MeV (fig. 3). The most important chain, the pp-chain, produces three kinds of neutrinos:

$$(1)\quad \text{PPI:}\qquad p + p \rightarrow d + e^+ + \nu\,.$$

The endpoint energy of the continuous pp neutrino energy spectrum is 0.42 MeV. pp-neutrinos can not be detected with a Chlorine detector (threshold 814 keV).

As already mentioned, more energetic neutrinos are produced in the two other branches of the pp-chain:

(2) PPII: $^{7}\mathrm{Be} + e^{-} \rightarrow {}^{7}\mathrm{Li} + \nu$,

(electron capture of ^{7}Be formed from ^{3}He + ^{4}He). These monoenergetic neutrinos (0.86 MeV) are only marginally detectable with the Chlorine detector. The expected flux of ^{7}Be-neutrinos is ~5 x 10^{9} $\mathrm{cm}^{-2}\mathrm{s}^{-1}$, it depends strongly on T_c ($\sim T_c^{8}$).

(3) PPIII: $^{8}\mathrm{B} \rightarrow {}^{8}\mathrm{Be} + e^{+} + \nu$,

(positron decay of ^{8}B formed from ^{7}Be+p). The endpoint energy of ^{8}B-neutrinos is 14.1 MeV. The expected flux is ~5×10^{6} $\mathrm{cm}^{-2}\mathrm{s}^{-1}$. It depends excessively on the central temperature ($\sim T_c^{18}$) hence the exact value of this very low flux is rather model dependent. Nevertheless, as already mentioned in principle it is possible to detect ^{8}B neutrinos with a Chlorine detector.

In addition, some neutrinos are also produced in the CNO cycle but in stars of one solar mass their fluxes are rather low, other than in hotter more massive stars.

In order to deduce expected production rates in a potential solar neutrino detector, John Bahcall also scrutinized the detector capture cross sections (*ft*-values for inverse beta decay) for the candidate target nuclides and estimated possible detection efficiencies for the product nuclides [2]. All partial neutrino branches, *i*, above threshold contribute to the signal as $S = \Sigma S_i = \Sigma\langle \Phi_i \sigma_i N_t \rangle$, where Φ_i is the neutrino flux, σ_i the cross section and N_t the number of target nuclei.

To obtain say 1 event per day, tens of thousand tonnes of material are needed, depending on the specific reaction. The appropriate unit for such minute production rates is the Solar Neutrino Unit (SNU): 1 SNU = one reaction per 10^{36} target atoms per second.

Concerning a Chlorine detector, the result was that even the weak ^{8}B neutrino channel could produce a few ^{37}Ar atoms per day in O(100 t) of ^{37}Cl.

There was also the question whether other astrophysical neutrino sources besides solar neutrinos that arrive at the detector could disturb the measured solar neutrino signal. However, this is not the case for the following reasons:

- Since the Sun is so much closer to the Earth than all other stars, the background in a solar neutrino experiment expected from fusion neutrinos of other stars in our Galaxy is completely irrelevant because of the power of $1/r^2$ scaling.
- A huge number of "Cosmological neutrinos" – with a flux of $O(10^{14}\ \mathrm{cm}^{-2}\mathrm{s}^{-1})$ on Earth – was produced in the Big Bang; yet their energy of $O(10^{-3}$ eV) is so low that they are undetectable and can be disregarded as potential background in solar neutrino experiments (in this range, the cross sections scale quadratically with the neutrino energy).
- Collapsing stars in the Galaxy that produce an intense neutrino burst with energies of O(10 MeV) are of course exciting research objects in their own, yet the Supernova-neutrinos contribute no background in solar neutrino studies since the collapses are rare and transient (typically lasting only for seconds). The $1/r^2$ effect also excludes any interference from the universal Supernova neutrino background.
- High Energy Cosmic Ray interactions in the upper Earth atmosphere produce high energy – O(GeV) – neutrinos (coined "atmospheric") that could eventually disturb measured solar neutron signals because of their higher interaction cross section. Yet their integral flux – $O(1\ \mathrm{cm}^{-2}\mathrm{s}^{-1})$ – is so low that their disturbance potential in spite of their high energy is at most marginal (<1%). Of much more practical concern in the context of radiochemical solar neutrino experiments are the cosmic rays themselves (nucleons, also as secondaries from deeply penetrating muons). They must be shielded to avoid (p,n) reactions on the target nuclide, a process that would mimic neutrino capture. Consequently, solar neutrino experiments must be accommodated in subterranean mines or mountain tunnels which are shielded by as much overlying rock as possible (thousands of meters). This rock should be low in natural radioactivities (U,Th,^{40}K) to prevent competing alpha or neutron-induced side reactions in the target.

It was not by chance that Bruno Pontecorvo had suggested the Cl-Ar scheme, because there were not many alternatives. For the radiochemical method to be applicable in practice, many prerequisites exist: The threshold has to be low, the lifetime of the product must not be overly short for effective accumulation (>few days) and not overly long to create enough specific activity (< few months). The isotopic abundance of the target nucleus must by sufficiently high. The chemical separation of the product from the target must be feasible with a separation factor of $O(10^{30})$. Ideal is Ar, a non-reactive volatile rare gas. A viable low-level counting method for the product nuclide must exist. Last but not least, multi-tons of the target element in sufficient purity must be accessible and affordable. A ^{71}Ga-^{71}Ge-Experiment (11.43 days half life of ^{71}Ge) was very desirable since the low threshold of 233 keV makes it sensitive to pp-neutrinos. However, in the early years the chances of realization for a Gallium experiment have been very bad because there was no worked out Ga-Ge separation procedure, no applicable counting technique for ^{71}Ge, and, in particular, no gallium. Tens of tons of the rare metal (twice the annual world production) were neither available nor payable.

On the experimental side, Ray Davis had operated a test experiment with 3800 liters of perchlorethylene (C_2Cl_4) close to the Savannah River Reactor and tested a He-purging

Fig. 4 The Homestake Chlorine Detector.

technique to quantitatively extract ^{37}Ar from large amounts of C_2Cl_4, admix it to the argon/methane counting gas and fill it into a gas proportional counter. The whole gas volume was kept below 1 cm^3. This allowed to miniaturize ultrapure gas proportional counters in order to lower the intrinsic counter background by orders of magnitude. The absence of a ^{37}Ar signal provided strong evidence for the difference between reactor (anti)neutrinos and solar neutrinos. The significant zero result encouraged Davis to conclude that the solar neutrino signal of a handful of ^{37}Ar atoms could in principle be detected and that backgrounds need not be prohibitive if the experiment were carried out in a laboratory deep underground.

In 1964, Bahcall and Davis published back to back two short notes in "Physical Review Letters" in which they summarized the status of their work up to this time [4]. From there on, solar neutrino detection changed from utopia to a feasible possibility.

In 2015, we celebrate the 50th anniversary of this landmark. The painful and joyful detail that followed was full of surprises. Skepticism of the scientific community remained dominant for a long time, yet at the end this new field of research led to fundamental insights far beyond the initial goal, namely the experimental proof that the life of the Sun is sustained by nuclear energy and continues to do so today. It culminated in the Nobel Prize that was awarded to Ray Davis in 2002. In this context personal reflections of mine on his personality and working style are presented in [5].

3 The Homestake Chlorine Experiment

It was clear that the detector had to be very big in order to produce at least a few neutrino induced ^{37}Ar atoms per month. At the same time, competing background reactions would demand preventive measures with very large depression factors. After a difficult search for a suitable site, Davis arranged with the management of the active Homestake Gold mine in Lead (South Dakota) the construction of an underground laboratory (fig. 4) shielded by 1480 m of rock, corresponding to 4200 m of water equivalent (m.w.e.). At that depth, the residual cosmic ray muon flux is reduced to ~4 muons/(d m^2). 380 000 liter of perchlorethylene (C_2Cl_4) were exposed in a single tank. This corresponds to 133 t of ^{37}Cl (the isotopic abundance of ^{37}Cl is 24.2%). For 5 SNU, one would expect that solar neutrinos produce about one ^{37}Ar atom/d, >10 times the known backgrounds to be expected. An exposure lasted typically for 2-3 months. Then, ^{37}Ar is flushed from the target in a helium stream together with some inactive carrier argon to trace the recovery. The argon is then collected at a charcoal trap, purified from non-inert gases and prepared for counting. For many years, Davis had tediously developed his ultra low background gas proportional counter ("Davis counter"). Special preparation techniques and extreme radiopurity of the counter materials led to backgrounds as low as <1 count per month in the ^{37}Ar signal acceptance window. After counter filling, counting lasted for 6–12 months in order to characterize also the counter background after the ^{37}Ar has decayed.

The signature of ^{37}Ar decay by electron capture back to ^{37}Cl is 2.8 keV Auger electrons. Rise time analysis of the point-like decay pulses is used to distinguish them from extended Compton-like background pulses.

Data taking with the Homestake detector started in 1968 and continued till 1995, results from 108 exposure periods ("runs") were collected. The overall result for this period was 0.48 ± 0.04 ^{37}Ar atoms per day. This corresponds to 2.56 ± 0.22 SNU (1σ), after subtraction of ~15% production due to known side reactions [5].

One should recognize that it is the statistics of more than 25 years of tedious operations that finally resulted in such a precise result, whereas the statistical significance of a single run is very close to zero. This is why this whole period of Homestake data taking was accompanied with claims of temporary fluctuations or periodicities that discredited the meaning of the results and therefore of the credibility of the questioned experiment altogether. Preoccupied super experts were just not willing to accept the fundamentals of the statistics of small numbers. Skepticism was frequent because of the unusual new experimental technique and the subtle nature of the signal, "playing billiard with a few atoms". In this critical phase, the institutional support or encouragement that Ray Davis received by BNL and, consequently, by DOE was rather limited.

Without the generous help of Ken Lande from Pennsylvania University, the Chlorine experiment would probably have come to an untimely early end. Consequently, Davis' working attitude was that of a lone wolf. In side experiments he could disprove virtually most intelligent and all not so intelligent suspicions. Unfortunately however, the plans to perform the ultimate performance test by exposing the Cl-detector to a suitable intense ^{65}Zn neutrino source did not materialize, mainly because of the above-mentioned problems and because of reactor-technological restrictions.

4 The Solar Neutrino Problem

The observed ^{37}Ar production rate of ~ 2.6 SNU made it highly probable that solar neutrinos had been observed for the first time. However, compared to the predicted rate of ~8 SNU, the observed rate corresponds to only ~1/3 of the expectation. This difference became known as the "Solar Neutrino Problem" (SNP). It seemed to indicate that the standard solar model does not fully describe the Sun and modifications are required that lower the ^{8}B-neutrino flux via a lower central temperature of the Sun.

Until ~1990, most attempts to explain this problem focused on details of solar physics. After 1990, the emphasis shifted from solar physics to particle physics as another possible cause of the missing electron neutrinos. This alternative interpretation was the possibility that we deal here with a propagation phenomenon in which something happens to the neutrinos on their way from the solar core to the detector on the Earth. On the way, they would in part change their identity so that they escape detection with the radiochemical detector.

It was already known that neutrinos come in three families (flavours) ν_e, ν_μ and ν_τ that are associated with electron, muon und tauon, respectively (in symmetry with the three existing quark families).

At the origin, solar neutrinos are electron neutrinos (ν_e) and radiochemical detectors are sensitive to ν_e only. As long as all neutrinos were massless, the flavours would not interact and a lepton number conservation law could exist for each flavour separately. However, if the flavour eigenstates had different masses, their flavour could oscillate during propagation along a travel path because of the velocity differences among the contributing mass eigenstates.

The principal conception of neutrino oscillations goes initially back to Bruno Pontecorvo, actually in that early stage he considered it for the case of neutrino-antineutrino oscillations. Going back to flavour oscillations, for long the preoccupation was that masses and mixing amplitudes should be – if not zero – then at least very small. Very small mass differences correspond to astronomical oscillation lengths, such as the Sun-Earth distance of 150×10^6 km. In 1978 it was recognized by Lincoln Wolfenstein, theoretician at the Carnegie-Mellon University in Pittsburgh, that high electron densities (*e.g.*, in the solar core) affect the mixing through coherent forward scattering of ν_e via charged currents. In 1984, Stanislav Mikheyev and Alexey Smirnov pointed out that the mixing amplitudes could be resonantly enhanced up to maximum mixing (level-crossing) in media of changing electron densities, even if they were small in vacuum ("MSW-effect").

This scenario exactly applies to our case: the solar neutrino beam propagates in a number of distinct environments: first the dense solar core, next the region of decreasing densities towards the solar surface, third the long distance in "vacuum", and finally the interior of the Earth (if the detector is at the night side). So the Sun is a very strong low-energy neutrino source that is well suited to test neutrino flavour oscillations and thereby to establish a non-vanishing neutrino mass. In short, the open question was whether the Solar Model or the neutrino mass is at the root of the Solar Neutrino Problem [2].

With the advent of the "Solar Neutrino Problem", a true symbiosis between neutrino particle physics and neutrino astrophysics appeared at the horizon. Nowadays, Astroparticle Physics has become a booming discipline and competes at the frontier line of particle physics on equal terms with high energy accelerator physics. The key question boiled down to whether there is a non-vanishing mass of neutrinos. This would imply that the Standard model of particle physics and interactions is not complete but "New Physics" is appearing at the horizon. The Solar Neutrino Problem started it all. The Chlorine experiment gave hints in this direction, but the uncertainties of the ^{8}B neutrino flux predictions and the missing neutrino source calibration made it impossible to allow a claim of such fundamental consequences. However, the hunt was on!

5 The GALLEX experiment

Detection of the most abundant but low-energy pp-neutrinos should lead to a clear distinction between the astrophysical and the particle physics solution of the SNP. The pp-neutrino flux is so accurately predicted from first principles explaining the well-known solar luminosity that an undisputed significant experimental deficit would make electron neutrino disappearance between Sun and detector the only viable explanation. The only (semi)practical option to measure pp-neutrinos was the possibility to use the reaction ^{71}Ga(ν_e, e$^-$)^{71}Ge with a threshold energy of 233 keV. However, a realistic experiment would require 10–100 t of gallium. In the late seventies and eighties of the last century, this demoralizing condition slowly changed from impossible to being a financial problem because of the boom of the aluminum industry in the eighties of the last century. Gallium occurs as trace element at the ppm level in bauxite, the principle ore for aluminum production. In such low concentration, it does not affect the metallurgical properties of aluminum and is not normally removed in the aluminum production process. However, if the aluminum industry had found it financially attractive to extract the gallium traces from the bauxite before aluminum production, theoretically there would have been now enough raw material to produce a few tens of tons of gallium within a few years. However, this would require the construction of specific gallium plants for the separation, the prize estimate for high purity gallium was in the range of 1–2 million dollars per ton, with all risks on the customer. There were many open questions concerning the realization of such a project:

- Could industry achieve the required radiopurity of the product?
- Would it be possible to develop a functioning Ge-Ga separation technique with a separation factor of $>10^{30}$? (in this respect, Cl-Ar separation is child´s play)
- Could one develop a Low-Level-Counting procedure for ^{71}Ge counting?
- Could one establish a committing international network of top scientists with the respective expertise and support by their agencies?
- Was there a suitable underground laboratory?
- Before all these questions are answered, could one dare to ask for funding of order 100 million dollars?

From 1966 onwards, the author of this narrative (Till Kirsten) was a postdoc and young Research Associate at the Chemistry Department of Brookhaven National Laboratory and worked with Ray Davis and Oliver Schaeffer on single atom detection of radioactive and stable atoms that were produced in natural cosmic-ray-induced reactions in extraterrestrial meteorites and, for interpretation, in the accelerator irradiation of specific target elements to determine high energy proton-induced spallation production yields. These data were wanted to interpret the time variation of cosmic rays from the records of their "cosmogenically" produced interaction products (radioactive and stable) in

Fig. 5 Ray Davis and Till Kirsten.

freshly fallen meteorites. While the author´s main expertise came from the mass spectrometric detection of extremely small quantities of stable rare gas isotopes (*e.g.* ^{128,130}Xe and ^{82}Se for Double Beta Decay Detection; ^{3}He, ^{36,38}Ar, ^{20,21,22}Ne for cosmogenic nuclide production), Ray Davis devoted all his efforts to push down the background level of gas proportional counters and to develop a low level counter to measure ^{3}H (tritium) and ^{37}Ar, clearly with solar neutrinos in the back of his mind. From there on he addicted me to the solar neutrino hunt (fig. 5). This was at the root of a life long lasting collaboration between BNL and the Heidelberg Max-Planck Institute for Nuclear Physics (MPIKH). The next few years were dominated by our institutes engagements in the NASA Apollo Lunar Sample Analysis Program in which our experimental capabilities of low level counting and determination of minute quantities of stable rare gas isotopes by mass spectrometry were urgently requested. We did our lunar job rather well, yet clearly Ray Davis considered this only as a training phase for a Chlorine detector. In my case I asked myself: “if men can go to the Moon and bring back samples that we analyze, why can‘t we get these damned 30 tons of gallium?”

So in 1979, we investigated the technical, fiscal and practical possibilities for a joint MPI/BNL Gallium experiment in the Homestake mine (fig. 6). Unfortunately, these efforts failed in all logistic aspects:

- The funding efforts at DOE failed in spite of John Bahcall´s influential help, most probably because the whole conception of radiochemical neutrino experiments had the image of being exotic, at best. More often, it triggered late party amusement at conference banquets.
- There was no additional space and infrastructure at Homestake. Quite understandably the much cheaper Chlorine experiment was closer to the heart of Ray Davis. In this situation, he chose to operate “low key” together with very few collaborators, primarily Bruce Cleveland and Keith Rowley.

A similar approach was out of question for a Gallium experiment. It was clear that a large international collaboration would be indispensable to produce and fund the required gallium and to solve all technical problems. This started our efforts to perform the Gallium experiment in Europe (fig. 7). This decision has never disturbed the friendly and fruitful cooperation between Heidelberg and Brookhaven during all coming years, Brookhaven’s radiochemists Robert Hahn and Keith Rowley have been important members of the GALLEX collaboration from early on.

Tests, pilot experiments and feasibility studies concerning ^{71}Ge low level counting in Heidelberg had already started in 1979. With great support from Antonino Zichichi, Nicola Cabibbo, Enrico Bellotti and Luciano Paoluzi, INFN set aside the required space and provided infrastructure in the Gran Sasso Underground Laboratory (LNGS), the first large underground facility that was exclusively devoted to fundamental research. GALLEX was the first experiment to operate there.

On commercial terms, Rhone Poulenc constructed the factory to produce 30 t of gallium after funding was assured by the Max-Planck Society and by the Alfried Krupp von Bohlen and Halbach Foundation.

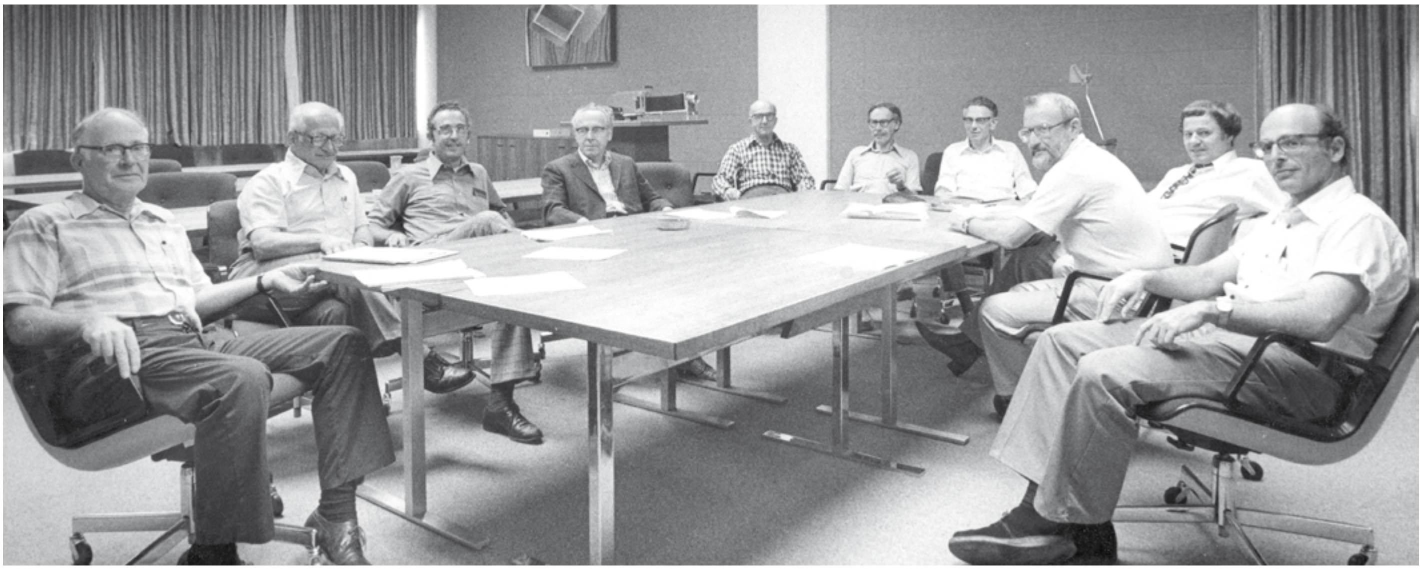

Fig. 6 Planning the BNL-MPI Gallium Experiment at BNL in 1979: clockwise from left: Ray Davis, Gerhard Friedlander, John Bahcall, Maurice Goldhaber, Israel Dostrovsky, N.N., Seymour Katcoff, Jerry Hudis, Till Kirsten, Ken Lande.

Fig. 7 GALLEX International Collaboration (France, Germany, Italy, Israel, USA).

Fig. 8 The GALLEX target tank.

Klaus Ebert and Edmund Henrich from FZK Karlsruhe contributed the large scale chemical engineering for the Ge-Ga separation under the difficult condition of radiochemical hyperpurity. It was decided to use the target in form of an aqueous solution of 8 molar gallium chloride (100 t, containing 30.3 t of gallium). In this highly acidic medium, the neutrino produced ^{71}Ge together with some inactive Ge carrier forms volatile germanium tetrachloride ($GeCl_4$) that is purged with nitrogen from the target solution. Later, the sample is chemically converted in GeH_4 that has favorable properties for gas proportional counting (in analogy to methane, the most preferred counting gas). In a large group effort at MPIK Heidelberg, Wolfgang Hampel and Gerd Heusser had developed the specific low level techniques for ^{71}Ge proportional counting at world record low background rates[3].

Construction and test operations in the LNGS lasted from 1986 to 1991. On May 14, 1991 the first Solar Neutrino recording started [3, 6, 7].

The GALLEX target tank (fig. 8) contained 100 t of 8 molar gallium chloride solution, equivalent to 30.3 t of gallium. This mega-sized teflon lined plastics tank was a technological novelty at the time. During 16 years of heavy operation, not a single drop of acidic liquid has ever leaked out. The tank was still fully intact when its further use was terminated by a spill elsewhere. This was an important experience for engineering in the chemical industry worldwide.

The production rate expected from the SSM was 130 SNU, corresponding to 1.16 ^{71}Ge atoms per day. This expected rate (see lower part of fig. 3) is the sum of 75 SNU from pp-neutrinos (PPI), 34 SNU from ^{7}Be neutrinos (PPII), 12 SNU from ^{8}B neutrinos (PPIII), and 9 SNU from neutrinos produced in the CNO cycle.

The 11.43 day half-life of ^{71}Ge defines about a month as the appropriate exposure time to approach saturation. At the end of each monthly exposure the tank was flushed with 3000 m^3 of ultrapure nitrogen gas for 12 hours to wash out the volatile germanium chloride for later clean lab bench chemistry and counting. These exposures lasted from May 1991 till January 1997, altogether 65 runs were executed. Figure 9 displays the continuous reduction of the statistical error of the GALLEX result during these 6 years, compared with the results of SAGE (see below).

Extremely important for the credibility of the GALLEX data was the verification of the whole experimental procedure by inserting a Mega Curie ^{51}Cr-neutrino calibration source into the GALLEX tank (fig. 10) and to perform ^{71}Ge-runs in full analogy to solar runs. When ^{51}Cr decays by electron capture with 28 d half-life in ^{51}V it emits a 746 keV neutrino. It can be produced by neutron activation of ^{50}Cr, but the production of a Mega Curie source was a formidable technological challenge. Rudolf Mößbauer from the Technical University

Munich (TUM) had joined the GALLEX Collaboration already in 1984. He used his good relations with the Kurchatov Institute in Moscow to arrange for the acquisition of 40 kg of chromium isotopically enriched in ^{50}Cr to 38.6% (the natural isotopic abundance of ^{50}Cr is 4.35% only).

The French CA Saclay group led by Michel Spiro and Michel Cribier took responsibility to neutron activate this enriched chromium at the SILOE reactor in Grenoble and to produce the GALLEX Mega Curie ^{51}Cr-neutrino calibration source, the strongest neutrino source ever produced. Two ^{51}Cr neutrino source Gallium irradiation experiments were performed in 1994 and 1995 and verified the proper operation of GALLEX. An additional performance verification ("^{71}As-test") was done in 1997 by adding a few hundred atoms of ^{71}As to the gallium chloride target, where they decayed into ^{71}Ge and were subsequently measured.

In 1998 the GALLEX collaboration changed to a new organizational structure in order to adapt to a more routine-like Gallium Neutrino Observatory (GNO). This was mainly an administrative adjustment whereby the essential experimental conditions remained basically unchanged. GNO data taking lasted from May 1998 to April 2003. Altogether, GALLEX and GNO have collected 10 years of neutrino data in 108 runs.

The historical *first* observation of pp-neutrinos was announced at the Neutrino 92 conference in the Alhambra gardens of Granada based on the first 15 runs. The result, 83 ± 21 SNU (1σ) implied a definite contribution from pp-neutrinos and thus, their discovery [7]. This converted "what nobody doubted to know about how stars produce their energy" into an experimental fact. Alvaro De Rújula, the summarizer of the conference quoted that the "solar pp-neutrino fusion bomb detonated over Granada by TK at 6.15 p.m. June 8th, 1992 ". In the foreword of the conference proceedings, Angel Morales judged that "the first GALLEX results will mark a cornerstone in Neutrino history. The participation in it of various historic scientists of neutrino Physics will prevail in our memory". This included Fred Reines who was the first to detect a neutrino, Bruno Pontecorvo who was overjoyed to see his visions come true, David Schramm, and many other celebrities.

Ironically, this positive observation of pp-neutrinos (an everlasting cornerstone of modern astronomy) became overshadowed by non-objective wishful thinking of many to prematurely interpret the deficit relative to the SSM expectation value (83 ± 21 SNU *vs.* 127 ± 20 SNU) as matter of fact indication of neutrino mass. A neutrino shortage from either pp or ^{7}Be neutrinos was evidently indicated, but the significance was <2σ. The GALLEX collaboration discussed the issue in depth [7] but refrained from a respective claim in the professional spirit that claims of seminal importance must be >3σ at least. In the following years GALLEX worked

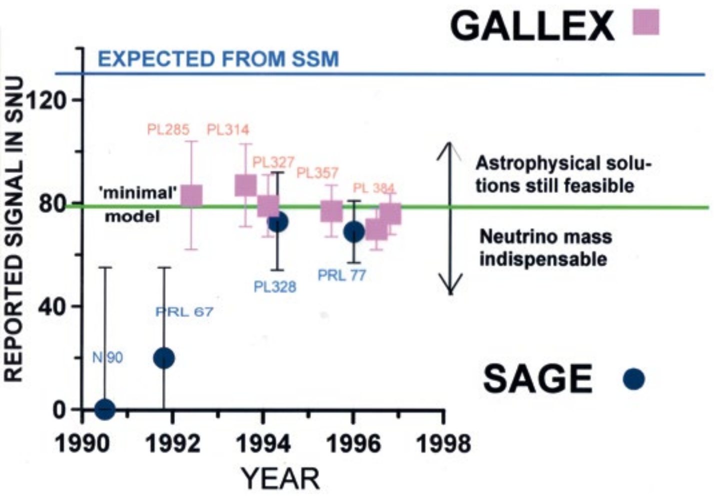

Fig. 9 GALLEX data 1992-1997. SAGE data for comparison.

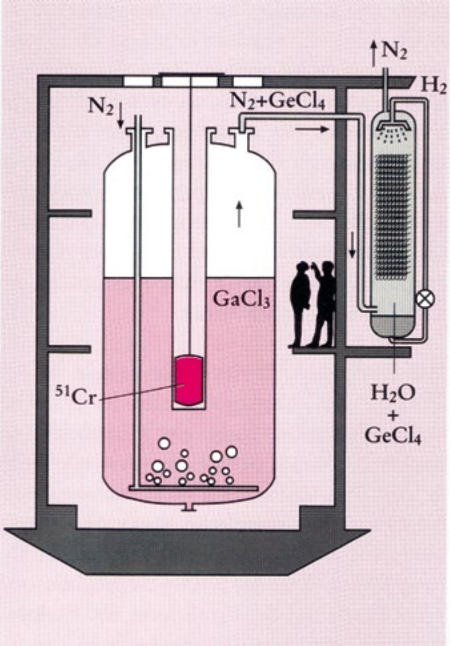

Fig. 10 Scheme of the ^{51}Cr neutrino source experiment to demonstrate the proper performance of GALLEX.

hard to replace unsupported premature claims by improving statistics through adding more and more runs. Consequently, the significance for the mass-mediated electron neutrino disappearance developed with improving statistics.

At the end of GALLEX data taking after 65 runs in 1997, the result, 77.5 ± 7.7 SNU was more than 6σ below the SSM prediction. It was significantly below a hypothetical minimal value that one obtains by simply adding the pp-flux requested from the solar luminosity and a ^{7}Be flux not higher than what is needed to account for the ^{8}B-neutrino flux (via the $^7\mathrm{Be}(\mathrm{p},\gamma)^8\mathrm{B}$) reaction in the Sun) that is measured in the Homestake Chlorine experiment. Hence, in 1997, disappearance of bulk (sub-MeV) neutrinos was finally assured at >99% confidence level. The acceptance of this result was further increased by the two rigorous performance tests already mentioned above: ^{51}Cr neutrino source exposures and ^{71}As spiking. Together, they established the long disputed reliability and reproducibility of the radiochemical method for neutrino detection in general and of GALLEX in particular. It was this final assurance of the reliability and significance of the pp neutrino flux deficit that was the first undisputed experimental evidence for "New Physics" [3].

6 The SAGE experiment

Parallel to GALLEX, another Gallium experiment has been performed by a Soviet-American collaboration in the underground laboratory at Baksan Valley, Caucasus. The *spiritus rector* of this experiment was Georg Zatsepin from the Russian Institute of Nuclear Research (INR), project manager was Vladimir (Volodja) Gavrin. The American contribution consisted predominantly in the generous help of Ray Davis in the provision and advice to operate his competitively low background gas proportional counters.

In this experiment, gallium was used as metal, liquid at 30 °C, in multiple containments ("reactors"). Here, the separation of germanium from gallium is achieved by mixing the metallic gallium with an aqueous solution that is acidic (HCl) and oxidizing (H_2O_2). A germanium atom will enter the aqueous phase if contact is made at the surface of a metallic gallium droplet. After germanium is in the acidic phase, the further chemical and counting procedures are in principle similar to those used in GALLEX.

Other than in the classical ion chemistry system of an aqueous solution that was applied in GALLEX, the SAGE separation technique was based on unexplored surface chemistry in a heterogeneous two-phase emulsion. This involved an unknown risk of unknown Ge-withholding mechanisms, in an experiment dedicated to operate at a few atoms level. Furthermore, substantial quantities of new chemicals had to be added in many charges at each run, thus making it difficult to ensure the radiochemical purity of the target and the uniformity from run to run. These are the objective reasons why skepticism of the scientific community prevailed in evaluating SAGE data releases before a convincing demonstration of systematic errors in such a completely new technology had been experimentally assured.

SAGE started solar neutrino recording in January 1990 and communicated a nil result after 6 month, this was advertised as indication of New Physics, but the claim was not justified in respect to statistical, and more importantly, systematical errors of an unproved procedure. The first serious data release covered the 6 early runs plus, after a one year interruption of no data or data rejected for unknown reasons, data from 8 further runs performed from June 1991 June 1992. This combined data set of 14 runs (specified as "SAGE 1") was quoted as 81 ± 20 SNU (1s), a few weeks after the practically identical GALLEX I result was released in Granada and "Physics Letters" (see fig. 10).

SAGE continued its recordings for many years and since 1996, perfect agreement between GALLEX/GNO and SAGE results was established. SAGE is continuing its measurements even today. On the contrary, the GALLEX/GNO operations were terminated in 2004 [8].

7 Subsequent solar neutrino physics with real-time detectors

With the progress of radiochemical neutrino experiments and the shift of priority from solar astrophysics to neutrino particle physics, the overwhelming accelerator physics community did not sleep. Given their superior financial possibilities (compared to the poor underground mules) they could push the technology of real time experiments by planning experiment sizes that where previously considered unrealistic. So experiments for the direct observation of incoming low-energy astrophysical neutrinos impinging on a detector were designed to become potentially competitive. This was accompanied by an ideological clash of philosophies on how best to solve the common problem of discriminating between the low rate signal – that is unchangeably provided by nature – and all kinds of backgrounds. The fantastic coincidence-anticoincidence techniques familiar to accelerator physicists tend to fail if rates and backgrounds approach the one per day or one per month level. The productive learning process between accelerator physicists and their colleagues that specialize in low-level techniques has well progressed during 30 years, but it is still not completed. The credo of low level physics is that the best way to veto background is to avoid it. Veto's are fine and necessary for the last step, but monitored and proved hyperpurity in materials and procedures are mandatory to begin with.

The first real time solar neutrino experiment was the 2100 t (680 t fiducial) water Čerenkov detector Kamiokande that started to operate in the Kamioka coal mine in Japan in 1987. It was based on neutrino electron scattering $e^-(\nu,\nu)e^-$. The recoil electrons are observed in forward direction relative to the position of the Sun. Limited by the high detector background at low energy, the energy threshold was > 7 MeV, hence only ^8B neutrinos could be observed. Nevertheless, this allowed a first comparison with the ^8B neutrino flux results from the Davis Chlorine Experiment.

The driving leader of Kamiokande was Masatoshi Koshiba. At Kamioka, he had daringly constructed the first large water Čerenkov detector to search for proton decay. In 1986, with no proton decaying, he decided to convert the detector from a "dirty" GeV experiment into a "clean" 10 MeV instrument to search for astrophysical neutrinos by lowering the intrinsic radioactive contamination of the water. Just months after this conversion was completed, Koshiba was blessed by heaven: it sent the Supernova 1987a, and Kamiokande observed the associated neutrino pulse. This opened him the Japanese financial channels for a very competitive Japanese astroparticle physics programme and later earned him the Nobel Prize 2002 together with Ray Davis, not for the solar neutrinos but for the Supernova.

With data taking starting in 1996, Kamiokande was drastically upscaled to SuperKamiokande with a fiducial volume of 22.5 kt. Under the guidance of Yoji Totsuka and Yoichiro Suzuki, this boomed the daily rate of ^8B neutrino events from O(1) to O(10), all registered with full kinematical and directional parameters [9]. Important for the success of the SuperKamiokande detector was the improvement of the vertex resolution of neutrino events through the advanced new timing electronics for each individual photomultiplier that was contributed by the US group led by Alfred K. Mann from the University of Pennsylvania.

With ongoing registrations of Homestake, Kamiokande, Superkamiokande, GALLEX/GNO and SAGE, errors shrank and the combination of the allowed parameter space in the plan of mass differences and mixing angles for each of those experiments led to a relevant picture of solar neutrino spectroscopy ("Large Mixing Angle Solution", LMA). For a more recent review see for instance [10]. The theoretical developments concerning neutrino oscillation scenarios improved the refinement of data interpretation to resolve in detail the everlasting conflict of solar model adaption's *versus* neutrino oscillation scenarios to explain apparent conflicts among the available experimental results. The final Rosetta stone concerning the role of neutrino mass in the solar neutrino story was contributed by the Sudbury Neutrino Observatory (SNO) experiment at the Creighton Nickel mine in Sudbury, Canada. It was designed and planned by Art McDonald from Queen's University in Kingston. Data taking started in 1999. The Čerenkov detector used 1000 t of heavy water (D_2O) in a transparent acrylic vessel to detect ^8B neutrinos above 5 MeV .

SNO is not restricted to Neutrino-Electron scattering: a) $\nu + e^- \rightarrow \nu + e^-$, but can also measure the flux and spectral shape of ^8B electron neutrinos (>5MeV) in the Charged Current (CC) reaction: b) $\nu_e + {}^2H \rightarrow 2p + e^-$ (^2H = deuteron).

However, the most attractive feature of SNO is the additional possibility to determine the total neutrino flux of all flavours, since the disintegration of the deuteron in the Neutral Current (NC) reaction: c) $\nu_x + {}^2H \rightarrow n + p + \nu_x$ is independent of neutrino flavour (x stands for all flavours: e, μ, τ). The neutron is detected by capture on ^{35}Cl from added salt or by ^3He counters.

The ratio CC/NC interactions yields the flux ratio $\Phi(\nu_e)/\Phi(\nu_{all})$. This searches for neutrino oscillations not only through the disappearance of ν_e but in effect also through the appearance (or reappearance) of ν_μ and ν_τ.

The satisfying result of SNO was that the total ^8B neutrino flux measured with the NC reaction agrees with the SSM expectation value, while the ν_e flux measured with the CC reaction agrees with the results from Kamiokande, SuperKamiokande, and Homestake. This was the final confirmation that solar neutrinos undergo flavour oscillations on their way from the solar core to the detector on the Earth. Further refinements in SuperKamiokande and SNO delivered a wealth of information on detailed neutrino properties, mass eigenstates and mixing parameters.

But how about the Sun? All available real-time data still concerned only those elusive rare ^8B neutrinos, the smell of the soup, but not the soup itself. So far, the only information about the soup itself still came from the radiochemical Ga experiments. The desire to apply the technological progresses in fast electronics and computing speed and at the same time adapting the benefits of low-level techniques led Gianpaolo Bellini from the University of Milano to try against all odds to built a real-time detector that can penetrate below 1 MeV, the range of ^7Be (0.8 MeV) and pp (<0.42 MeV) neutrinos. This is the BOREXINO liquid scintillator experiment (see fig. 11) installed in the hyperpure pseudocumene organic scintillator contained in a thin transparent nylon vessel that is observed by 2200 photomultpliers to detect neutrino scattering on the scintillator electrons. One obtains a full spectrum from the various overlapping neutrino and background sources. In this way it became possible to identify the 862 keV ^7Be-neutrino flux, the ^8B neutrinos (down to 3 MeV), the 1.44 MeV "pep neutrinos" [11] and, just recently, even

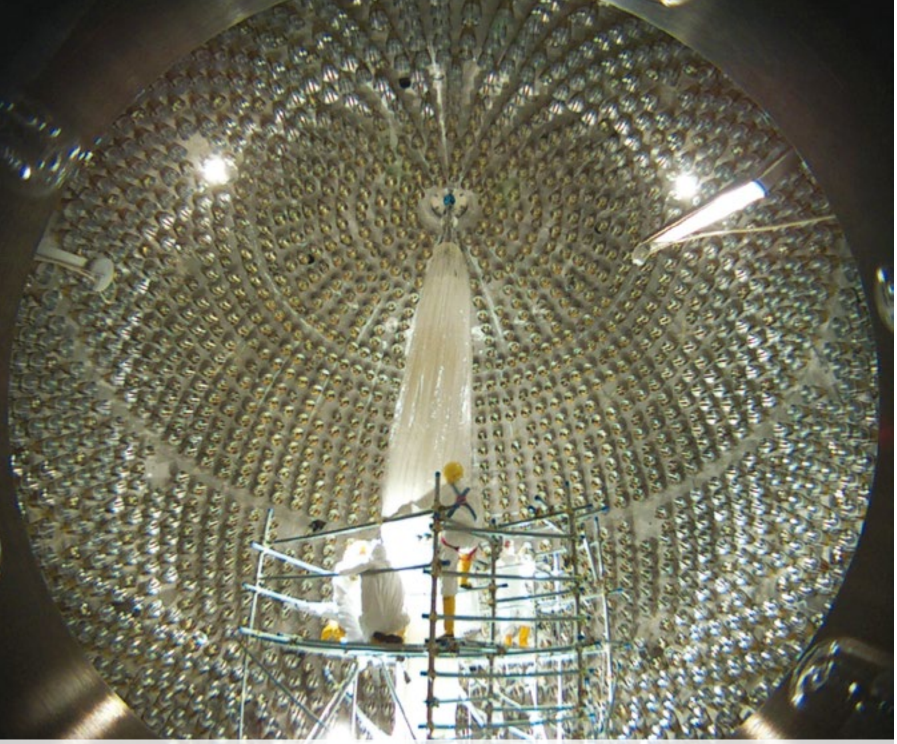

Fig. 11 Inside the BOREXINO with the photomultipliers arrays, while installing the inner baloon. Borexino Collaboration-LNGS/INFN.

the edge of pp-neutrinos. This closes (for the moment) the circle of discoveries that was initiated by the radiochemical experiments.

Today we know the main features of the solar neutrino spectrum as well as the spectrum of masses and mixing of neutrino mass eigenstates. Rare event physics is now a flourishing field in the further exploration of neutrino properties such as the distinction between Majorana and Dirac neutrinos, as well as searches for neutrinoless Double Beta decay and for Dark Matter. Solar model refinements concerning its metallicity, and even geophysical probing of the interior of the Earth globe using neutrinos from terrestrial radioactivity round up the spectrum of modern neutrino physics. Neutrinos are no longer elusive, at present they are even used for nuclear non-proliferation surveillance. A century (±2%) after Bruno Pontecorvo was born in Pisa, a creative thinker ahead of his time, unspoiled by his personal vita.

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Till A. Kirsten received the PhD. in Physics at the Heidelberg University in 1964. Professor of Physics at Heidelberg University since 1975, he has been in the Max-Planck Institut für Kernphysik (MPIK) in Heidelberg since 1972, with which he maintains his association after his retirement in 2002. In 1969 he received the Röntgen award for the experimental proof that double beta decay occurs in Nature. From 1974 to 1984 he was the Principal Investigator in the NASA Apollo Lunar Sample Analysis Program. In 1985, he initiated and subsequently led the GALLEX experiment for solar pp-neutrino detection at the INFN LNGS. In 1992, the experiment succeeded in the first direct observation of hydrogen fusion in a star. For this, Kirsten received the Gentner-Kastler Prize in 1993. He subsequently worked in the Gallium Neutrino Observatory, from 1997 to 2003, and in the BOREXINO solar neutrino experiment since 1995, both at the LNGS.