## Solar neutrinos: the pioneering experiments

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The neutrinos that are produced in the solar fusion reaction chains reflect the conditions in the stellar interior. If the model predictions for the expected neutrino fluxes are firm, then the solar neutrino experiments can also test neutrino properties during propagation along the 150 million km 'baseline' between the 'source' and the detector. After the initial visionary conception of Bruno Pontecorvo in 1946 for a <sup>37</sup>Cl - <sup>37</sup>Ar detector based on the radiochemical accumulation principle, experimental efforts by Raymond Davis jr. started around **1967**. Here I cover mainly all relevant experimental activities in the subsequent  $\approx$  **30** years, in a historical context. Results during this period have been acquired with 4 solar neutrino experiments: Homestake, Kamiokande, Gallex, and Sage. Major milestones achieved between 1970 and 1996 have been:

- 1970 (-1994): first successful application of the **radiochemical** detection method (Homestake Chlorine detector) to measure solar neutrinos. Detected were mainly <sup>8</sup>B neutrinos from the PPIII chain with energies > 814 keV.
- 1987: first detection of <sup>8</sup>B neutrinos in a **realtime** experiment (Kamiokande water Čerenkov detector, sensitive to PPIII chain > 7.5 MeV). Confirmation of the <sup>8</sup>B neutrino deficit that was observed in the Chlorine experiment ('Solar Neutrino Problem').
- 1992: first observation of hydrogen fusion in the solar interior by positive detection of solar ppneutrinos with the radiochemical Gallium detector at the Gran Sasso Underground Laboratory in Italy (Gallex). Detected were solar neutrinos mainly from the PPI and PPII chains with energies > 233 keV. At the same time, a definite deficit of pp-and/or <sup>7</sup>Be neutrinos was established.
- 1995: first **assurance** of non-zero neutrino mass (most probably related to neutrino flavor oscillations) after Gallex III and the <sup>51</sup>Cr neutrino source experiments.

The 'pioneering phase' of establishing new experimental methods and of initial discoveries ended with the advent of Super-Kamiokande in 1996, beginning a new phase of systematic astroparticle physics that is however, not subject of this article.

## 1 Solar Model and Neutrino Fluxes

The solar model for the prediction of solar neutrino fluxes describes the state of the stellar interior based on hydrostatic equilibrium, the (ideal gas) equation of state, radiation dominated thermal equilibrium, and energy production by hydrogen fusion, associated with neutrino production. Major input data are the solar mass, radius, luminosity, age, chemical composition, S-factors (nuclear reaction cross sections) and radiative opacities.

Solar models have been continuously elaborated and improved from the earliest times of Eddington, Critchfield and Bethe till today, with base setting contributions particularly by John Bahcall since  $>1960^{1,2,3}$ . The solar fusion reaction chains are shown in Figure 1.



Figure 1 – Solar fusion reaction chains.

The chains are associated with the production of pp (and pep) neutrinos (PPI); <sup>7</sup>Be-neutrinos (PPII) and <sup>8</sup>B-neutrinos (PPIII). Also listed are the branching ratios for pp- <sup>7</sup>Be-, and <sup>8</sup>B-neutrinos. The spectrum of these neutrino branches is shown in Figure 2, based on <sup>4</sup>.



Figure 2 – Solar neutrino spectrum.

The experimental challenge to measure these neutrinos is extremely ambitious because of incredibly small cross sections and excessive demands for background reduction. Obviously, the lower the energy of the neutrinos, the more difficult becomes the task to detect them.

The pp neutrinos are, by far, dominant in intensity while lowest in energy ( $\leq 420$  keV). The flux at the Earth is  $\approx 6 \times 10^{10} / \text{cm}^2$ s and it is robustly insensitive to the particulars of any

solar model variations. In fact, this flux follows directly from the solar luminosity according to the sum equation given in Fig.1. For the case of the <sup>7</sup>Be neutrinos, the flux at the Earth is predicted to be  $\approx 5 \times 10^9 / \text{cm}^2$ s and varies approximately with the  $10^{th}$  power of the central solar temperature. Their energy is intermediate, and they are mono-energetic line sources at 0.86 MeV and at 0.38 MeV.

The <sup>8</sup>B neutrinos are rare  $(5 \times 10^6 / \text{cm}^2 \text{s})$ , are model dependent  $(18^{th} \text{ power of the central temperature})$  and have a continuous energy spectrum up to  $\approx 14$  MeV. Suppression of this channel for energy production would have no impact on the solar luminosity.

The neutrinos leave the solar core virtually unhindered, distinct from photons or any other radiations. After penetration of  $\approx 700,000$  km of solar matter at decreasing density and after subsequent travel through interplanetary space for 150 million kilometers (1 AU), the neutrinos can reach a detector on Earth, either from the front or from the back, within  $\approx 8$  minutes, practically in real time. If detected, the neutrinos can serve as messenger particles about the present state of the solar interior and the reactions occurring therein. This allows experimental tests of theoretical solar models. Such knowledge is fundamental for astrophysics and for models of stellar structure in general since the Sun is a standard main sequence star<sup>*a*</sup>.

From the particle physics point of view, the Sun is a very strong low-energy neutrino source at a great distance. It is well suited to test the properties of a neutrino 'beam' while it propagates a number of distinct environments: first, the very dense medium in which the neutrinos formed, second, the region of decreasing electron densities that occur as the neutrinos travel to the solar surface, third the vacuum at space, and finally, the interior of the Earth if the detector is at the night side. These are ideal conditions for a 'longbaseline experiment' to search for neutrino flavor transmutations and thus for manifestations of a nonvanishing neutrino rest mass. For this application, the source strengths (the neutrino source function) must be reliably known, and the detector must be flavor specific. Solar neutrinos are generated as electron neutrinos  $\nu_e$ . A detector tuned to  $\nu_e$  would register a flux-deficit relative to expectation if transformations into muon- or tau-neutrinos occur (i.e., a 'disappearance experiment'), see Figure 3.



Figure 3 – Principle of solar neutrino flavor-oscillations.

### 2 The Radiochemical Method

The radiochemical detection technique approaches the problem of extremely low interaction rates by using very large target masses and by collecting the radioactive neutrino induced reaction products over extended time periods, typically a few half-lives of the product nuclei. After this conceptional impetus<sup>5</sup> by Bruno Pontecorvo (Figure 4), it was first applied by Raymond Davis

 $<sup>^{</sup>a}$ See a brief bibliography on the solar neutrino problem and on the Standard Solar Model (SSM) at the address https://neutrino-history.in2p3.fr/solar-neutrinos (end of that sub-section) and a shorter version on SSM in a dedicated page after this talk.

jr. (Figure 5) - for his Chlorine detector, in which, however, pp-neutrinos were inaccessible because their energy is below the threshold of the  ${}^{37}\text{Cl}(\nu_e, e^-){}^{37}\text{Ar}$  reaction.



Figure 4 – Bruno Pontecorvo.



Figure 5 – Raymond Davis jr.

The method is based on inverse beta decay (neutrino capture) of suitable target isotopes. The product nuclei  ${}^{A}(Z+1)$  from the neutrino capture  ${}^{A}Z$  ( $\nu_{e}$ , e<sup>-</sup>)  ${}^{A}(Z+1)$  are accumulated during exposure of large targets to near saturation. Then the reaction product is separated from the target with radiochemical techniques and subsequently detected by radioactive decay. Only the energy threshold of the selected reaction restricts the minimum energy for neutrino detection. Interfering side reactions can be kept in principle at a level that is negligible or at least acceptable after a respective numerical correction.

All partial neutrino branches i above threshold contribute to the signal  $S_i$  as  $S = \Sigma S_i = \Sigma \langle \phi_i \sigma_i N_t \rangle$  (1) where  $\phi_i$  = neutrino flux,  $\sigma_i$  = cross section,  $N_t$  = number of target nuclei. Required are  $E_{threshold} \langle 420 \text{ keV}$  for detection of pp-neutrinos,  $E_{threshold} \langle 862 \text{ keV}$  for <sup>7</sup>Beneutrinos, and  $E_{threshold} \langle 14.1 \text{ MeV}$  for <sup>8</sup>B-neutrinos.

For an experiment to be meaningful, it is necessary that reliable theoretical estimates are available for all these contributions. This involves the Standard Solar Model parameters, the relevant cross sections (S-factors) and the detector capture cross sections (ft-values for inverse beta-decay). Another critical condition is the reduction of production background. For this, both cosmic radiation and natural radioactivity must be reduced to far below the normal environmental level, since secondary protons ( $E_p>1$  MeV) can mimic neutrino capture via (p,n) reactions in both cases.

Cosmic rays can be shielded by going underground; all radiochemical solar neutrino experiments are located in deep mines or in mountain tunnels. An overburden of 1000 meters of rock corresponds to a shielding of 2500 - 3000 meter water equivalent (m.w.e.).

The radiochemical purity must be, depending on the experiment, in the range of  $10^{-10}$ - $10^{-16}$ g U, Th per gram of target because the  $\alpha$ -decay series interfere via ( $\alpha$ ,n) and (n,p) sequences. Another background source is (n,p)-reactions due to fast neutrons emitted from the rock walls of the underground lab. An important practical requirement is the feasibility of the chemical separation of the product nuclei from the target. Huge separation factors, up to  $10^{30}$ , are needed. Ideal are schemes where the neutrino capture product is volatile and can be flushed with a gas stream, as is the case in the Homestake- and in the Gallex experiments (see sections 3,5).

After the product nuclei are successfully separated from the target, they must be counted. The products from inverse beta decay are radioactive by electron capture; the detectable radiation in this process consists of keV-Auger electrons and X-rays. Such weakly ionizing low energy radiation is normally detected in low-level gas proportional counters which contain the radioactive species as an admixture to the counting gas. Typical decay rates are of order  $\leq 1/\text{day}$ . This calls for counter backgrounds  $\leq 1/\text{week}$ . Techniques towards achieving this goal are ultimate low-level procedures, radiochemical purity, counter miniaturization, as well as energy and pulse shape analysis with fast electronics (transient digitizer, neural network analysis).

Last but not least, the target material for a radiochemical solar neutrino experiment must be obtainable and affordable in sufficient quantity and purity.

#### 3 The Homestake Chlorine Experiment

Raymond Davis jr. at Brookhaven National Laboratory (BNL) was the first to demonstrate that it is indeed possible to detect a few atoms out of hundreds of tons of target material. In the late sixties, he constructed the first solar neutrino detector in the Homestake gold mine in Lead, South Dakota<sup>6</sup>. The aim was to detect mainly <sup>8</sup>B-neutrinos via the reaction <sup>37</sup>Cl( $\nu_e$ , e<sup>-</sup>)<sup>37</sup>Ar, as proposed by Bruno Pontecorvo. Large quantities of chlorine were conveniently available in form of perchlorethylene (C<sub>2</sub>Cl<sub>4</sub>) at reasonable cost. The half time of <sup>37</sup>Ar is 35 days.

The energy threshold for neutrino capture on  ${}^{37}$ Cl is 814 keV, far above the 420 keV maximum energy for pp-neutrinos. On the other hand, <sup>7</sup>Be-neutrinos can contribute to the production of  ${}^{37}$ Ar from  ${}^{37}$ Cl since their energy exceeds the threshold.

The theoretical expectation value for the  ${}^{37}$ Ar production in a chlorine detector has been relatively uncertain and varied between 6 and 10 SNU<sup>b</sup>. The rate for  ${}^{8}$ B-neutrino capture on  ${}^{37}$ Cl is dominated by the transition to the isobaric analogue state at 5.1 MeV. This allowed the determination of the transition strength from the properties of  ${}^{37}$ Ca beta decay.

Most of the uncertainty of the rate prediction for the chlorine detector is due to the uncertainty of the estimate for the <sup>8</sup>B-flux. It is extremely sensitive to even slight modifications of the solar model or of the input parameters used. Any change modifies  $T_c$ , the central temperature of the Sun, and the predicted <sup>8</sup>B- $\nu$  flux responds proportional to  $T_c^{18}$ . Another uncertainty stems from the relative insecurity of the <sup>7</sup>Be(p, $\gamma$ )<sup>8</sup>B cross section.

For the Standard Solar Model (SSM) with inclusion of heavy element diffusion, Bahcall et al.<sup>4</sup>) deduced a production rate for the chlorine detector of  $7.7 \pm 1.2 \pm 1.0$  SNU (1 $\sigma$ ). About 85 % of this rate is due to <sup>8</sup>B neutrinos,  $\approx 15$  % is expected to come from <sup>7</sup>Be neutrinos. The error is mainly due to the uncertainty of the contribution from <sup>8</sup>B-neutrinos (> 90%). About 100,000 gallons perchlorethylene were exposed in a single tank. This corresponds to 133 tons of <sup>37</sup>Cl (the isotopic abundance of <sup>37</sup>Cl is 24.2%). The expected production rate was  $\approx 1.5$  <sup>37</sup>Ar atoms/d.

The tank was shielded by 1480 m of rock or 4200 m.w.e. At this position, the residual cosmic ray muon flux is only  $\approx 4 \text{ muons/d,m}^2$ . The <sup>37</sup>Ar production from this source was estimated to be 0.05 <sup>37</sup>Ar atoms/d. All 'known backgrounds' together would contribute < 0.1 <sup>37</sup>Ar atoms/d.

Governed by the 35 d half-life of  ${}^{37}$ Ar, an exposure run lasted typically for 2-3 months. Then,  ${}^{37}$ Ar was flushed from the target in a helium stream together with some inactive carrier argon to trace the recovery. Argon is then collected on a charcoal trap, purified from non-inert gases, and admixed to the counting gas of miniaturized low-level gas proportional counters. For background reduction, the active counter volume was kept as small as  $\approx 1 \text{ cm}^3$ .

Counting lasted for 6-12 months in order to characterize the counter background after the decay of <sup>37</sup>Ar. The reduction of this background to the necessary level has been a major achievement. The signature of <sup>37</sup>Ar decay by electron capture is 2.62 keV Auger electrons. Low background for such low energy did require extreme radiopurity of the counter materials and special preparation techniques. Remaining background pulses have been mostly slowly rising Compton-like extended events. They are due to partial energy deposition of energetic particles that cross the active counter volume. These background pulses are distinguished from fast point-like <sup>37</sup>Ar-decays by rise-time analysis.

After all cuts for energy, rise time, and some other parameters, an event list is established. Next, the maximum likelihood method is used to partition the counts in a signal that is characterized by a 35-day half time and a background that is constant in time. Typical background rates achieved are less than one count per month; some counters are even better.

<sup>&</sup>lt;sup>b</sup>1 SNU = 1 Solar Neutrino Unit = 1 reaction per  $10^{36}$  target atoms and second.

Data taking with the Homestake detector started 1968 and continued till 1994, with only one serious interruption due to erratic developments of BNL policies yet fortunately resolved by Ken Lande (Pennsylvania State University). Figure 6 displays the results of all individual runs; the interruption is marked in yellow.



Figure 6 – Individual run results from the Homestake Chlorine experiment<sup>7</sup>.

A single run result has very little statistical significance since one observes only a few events per run. On the other hand, the overall distribution of these individual results is consistent with the assumption of a production rate constant in time. This justifies the deduction of a mean value. For the full data set, this yields a statistical error as small as  $\pm 6\%$  of the measured signal  $(1\sigma)$ .

The overall result is an <sup>37</sup>Ar production rate of  $0.484 \pm 0.042$  <sup>37</sup>Ar atoms/d. This corresponds to  $2.56 \pm 0.16$  (stat.)  $\pm 0.15$  (syst.) SNU or, with errors combined in quadrature,  $2.56 \pm 0.22$  SNU  $(1\sigma)^{7}$ . Side reactions (already subtracted) amount to  $\approx 15$  % of this rate. At 95 % c.l. the observed rate is  $\leq 3$  SNU, and this must be compared to the predicted rate,  $7.7 \pm 1.2 \pm 1.0$  SNU. The observation corresponds to  $(33 \pm 6)$  %  $(1\sigma)$  of this expectation.

The deficit became known as the 'solar neutrino problem' (SNP). At this time, it seemed to indicate that the SSM does not fully describe the Sun and modifications are required that lower the <sup>8</sup>B-neutrino flux by virtue of a lower central temperature of the Sun. Alternatively,  $\nu_e$  disappearance (see Fig.3) attained increasing attention. However, before such far-reaching options were supported, many objections and suggestions of potential experimental errors were raised. Ray Davis and collaborators could disprove virtually all such suspicions in side experiments and proved that the overall results of the chlorine experiment are reliable. For absolute assurance, the ultimate test of exposing the Cl-detector to a calibration source of low energy neutrinos was unfortunately never realized, to the large regret of Ray Davis.

## 4 The Kamiokande (real time) Experiment

Additional experimental data on solar <sup>8</sup>B neutrinos became available since 1987 from the Kamiokande water Čerenkov detector <sup>8</sup> based on neutrino electron scattering, e<sup>-</sup> ( $\nu_x, \nu_x$ ) e<sup>-</sup>. This detector was initially at the Kamioka mine in Japan to search for proton decay. In 1986, it was converted from a GeV-detector in a 10 MeV-detector. This was accomplished by lowering the intrinsic contamination of the water. The longlived natural radioactivity (U,Th decay series,<sup>40</sup>K) was reduced to a level at which it became feasible to observe the neutrino-induced recoil electrons having energies as low as  $\approx$  7 MeV. In this way, at least the upper part of the <sup>8</sup>B solar neutrino spectrum became accessible. The detector was installed below 2700 m.w.e. of shielding. This is not quite as much as one would ideally wish to have but it is enough to reduce

the background from muons and short-lived spallation products in the water to an acceptable level.

The detector can record data on the conic Čerenkov-light emission produced by recoil electrons in the 2142 tons of water using a dense network of PMT's and a 2-meter wide anticoincidence cover. The innermost 680 tons were used as the sensitive fiducial volume. The energy and the position of an event are reconstructed by using the number and geometric orientation of the PMT cells hit by the Čerenkov light cone together with the relative event arrival times. The energy resolution in Kamiokande was 22% at 10 MeV; the vertex resolution 1.7 meter; the angular resolution  $\pm 28^{\circ}$ ; and the time resolution 5 ns.

The cross section for  $\nu_e$ -e<sup>-</sup> scattering of solar <sup>8</sup>B neutrinos with >10 MeV is  $\approx 2 \times 10^{-45}$  cm<sup>2</sup>. The spectrum of the recoil electrons reflects the initial neutrino spectrum in a theoretically well-understood fashion. Triggered events are sorted according to the number N<sub>h</sub> of PMT's hit per event. For instance, a 10 MeV neutrino yields an average N<sub>h</sub> =22 in Kamiokande.

In spite the provisions described above, the task to recognize solar neutrino induced events was formidable. It required extremely good criteria for track recognition and timing. Note that the total trigger rate was 150,000/d, while the expected signal surviving all necessary cuts is < 1 event/d. The major cuts are based on selecting the energy acceptance window (e.g.,  $22 < N_h < 36$ ); rejecting muon-induced events with the outer anticoincidence counter, and setting the event separation time to >100  $\mu$ s in order to recognize decay electrons from stopped muons; requiring the vertex to be inside the fiducial volume in order to reject external  $\gamma$ 's and neutrons; performing a time analysis to reject 10-15 MeV beta particles from short-lived, muoninduced spallation products on oxygen; and finally, on requiring the event to be produced from the direction of the Sun, to within a  $37^{\circ}$  cone at a 90 % probability for acceptance. After all these cuts a signal from the Sun's direction is clearly visible. However, it is not nearly as high as predicted from the SSM. The mean observed event rate for the solar signal was found to be  $\approx 0.4/d$  during the 2079 days of Kamiokande observations between January 1987 and February 1995. The <sup>8</sup>Bneutrino flux measured by Kamiokande is  $(2.82 \pm 0.19 \text{ [stat]} \pm 0.33 \text{ [syst]}) \times 10^6/\text{cm}^2\text{s}^9$ . This is to be compared to the theoretical flux prediction of  $(5.15\pm0.98\pm0.72)\times10^6/\text{cm}^2\text{s}$ . In summary, only 55 % of the expected <sup>8</sup>B neutrinos are found. The deficit of 45 % is significantly larger than the errors of the flux measurement by Kamiokande. Cum grano salis, the  ${}^{8}B-\nu$  deficit in the Homestake detector was confirmed in a 'classical' real-time experiment.

## 5 The GALLEX (+GNO) Gallium Experiments

The measured signal in the Homestake experiment turned out to be only at a level of about 1/3 of what was roughly expected from the SSM for 8B-neutrinos. This established the SNP, see<sup>10</sup>). The deficit could have been caused either:

- by deviations due to an incomplete or false description of the solar interior by the SSM and/or by inaccurate input parameters: astrophysical solution of the SNP or:
- by non-standard neutrino properties: particle physics solution of the SNP (like, e.g. non-zero neutrino mass at the root of neutrino flavor oscillations).

If a significant deficit were observed for pp-neutrinos, one could rule out the astrophysical solution since the pp-flux at origin is directly fixed to the well-known solar luminosity. pp-neutrinos are by far the most abundant solar neutrinos, yet their energy is very low (<420 keV). This demands a detection reaction with very low threshold. The only practical option was  $^{71}$ Ga( $\nu_e$ , e<sup>-</sup>)<sup>71</sup>Ge, with a reaction threshold of 233 keV. This reaction scheme was first suggested by V.A.Kuzmin<sup>11</sup>. However, a realistic experiment would require 10-100 tons of gallium. In the late seventies and eighties of the last century, this was a demoralizing condition.

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 would find it financially attractive to extract the gallium traces from the bauxite before aluminum production, theoretically there was 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 M\$ 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 <sup>71</sup>Ge counting?
- Could one establish a committing international network of top scientists with the respective expertise and support by their agencies?
- Is there a suitable underground laboratory?
- Before all these questions are answered, can one dare to ask for funding of order 100 M\$?

At the root of a life long lasting collaboration in Cosmochemistry and Nuclear Chemistry between the 'Brookhaven National Laboratory' (BNL) and the 'Heidelberg Max-Planck Institut für Kernphysik' (MPIK), since 1979, Raymond Davis and Till Kirsten investigated the technical, fiscal and practical possibilities for a joint MPIK/BNL gallium experiment in the Homestake mine (see Figure 7). Unfortunately, these efforts failed because of lacking administrative BNLsupport, lack of funds and lack of underground space at Homestake or elsewhere in the US or in Canada. As a consequence, in 1983 T.K. started efforts to perform a gallium experiment in Europe. This decision has never disturbed the friendly and fruitful cooperation between Heidelberg and the Brookhaven colleagues during all coming years, Brookhaven's radiochemists Robert Hahn and Keith Rowley have been important members of the Gallex collaboration from early on.



Figure 7 – BNL-MPIK coordination meeting, March 1979. Clockwise from left: Ray Davis, Gerhard Friedlander, John Bahcall, Maurice Goldhaber, Israel Dostrovsky, N.N., Seymour Katcoff, Jerry Hudis, Till Kirsten, Ken Lande.

Tests, pilot experiments and feasibility studies concerning <sup>71</sup>Ge low level counting in Heidelberg had already started in 1979. With great support from Nicola Cabibbo, Luciano Maiani and Enrico Bellotti (Figure 8), 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.



Figure 8 – From left to right: Luciano Maiani, Nicola Cabibbo, Enrico Bellotti.

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

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 hyper purity. It was decided to use the target in form of an aqueous solution of 8 molar gallium chloride (100 tons, containing 30.3 tons of gallium). In this highly acidic medium, the neutrino produced <sup>71</sup>Ge together with some inactive Ge carrier forms volatile GeCl<sub>4</sub> that is purged with nitrogen from the target solution. Later, the sample is chemically converted in GeH<sub>4</sub> (germane) that has favorable properties for gas proportional counting (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 <sup>71</sup>Ge proportional counting at world record low background rates <sup>12</sup>.

Construction and test operations in the Gran Sasso underground laboratory (LNGS) lasted from 1986-1991. On May 14, 1991 the first Solar Neutrino recording started <sup>13,14,15</sup>.

The conception of the Gallex experiment is fully described in <sup>16</sup> and in many earlier (e.g. <sup>17</sup>) and later (e.g. <sup>18</sup>) collaboration papers. Here I will not repeat the details but restrict myself to the major characterization of principal technical aspects of the experiment:

- Target size: Limitations are financial costs and limited availability. We needed an annual world production of high purity gallium, at a prize of  $O(10^7 \text{CHF})$ . An extra Ga-factory (extraction from bauxite) was constructed for this by our supplier.
- <u>Ga-Ge separation</u>: The advantage of using acidic gallium-chloride solution is that any Ge will automatically form volatile germanium tetrachloride that can be purged out of the solution using hyper-pure nitrogen. This is later (in lab-scale) transformed in germane, GeH<sub>4</sub>, ideal for gas proportional counting (note the analogy to methane, CH<sub>4</sub>, our counting gas is Xe/GeH<sub>4</sub>). We use only very small quantities of stable germanium carrier for yield control.
- Proportional counting: The Ge-decay peaks occur at 1.17 keV (L-peak) and 10.37 keV (K-peak). Pulse shape analysis distinguishes between genuine fast rising pulses due to point-like <sup>71</sup>Ge decays and slow pulses of extended ionization tracks from Compton-like background events where the primary ionization occurs at various radial distances from the anode wire; for more detail see <sup>16</sup>).
- <u>Run sequence</u>: Typically, we performed one run (extraction) per month, governed by the favorable mean life of <sup>71</sup>Ge (16.49 d). Extraction time was  $\approx 1$  day, each run was counted for  $\approx 6$  months.
- <u>Purity philosophy</u>: In this type of low rate experiments (few counts per monthly run), radiochemical purity in every object and process, from acquiring the target material through every procedural step till completion of counting, ultra-low-level purity is of paramount

importance, and yet, it must be controlled at a level that is even more difficult to measure than the signal. Anticoincidence filter in counting are crucial for the last step, but they would be useless if the sample would be contaminated to begin with. The first way to reduce background is to avoid it! This is the idea behind the miniaturization of our proportional counters: 'less material, less background!'. In rare event physics, accelerator physics and low-level physics must come together, an educational difference of attitude is often encountered in large collaborations at the edge of these fields.

The conceptions of the experiment are compiled in Figure 9, illustrating sketches of some hardware components are depicted in Figure 10.



Figure 9 – Conceptions of the Gallex Experiment.



Figure 10 – Gallex components. Clockwise from top left: (i) arrival of one of the two 70 m<sup>3</sup> target tanks at the A24 highway tunnel entrance, 6.3 km from the Gran Sasso underground laboratory, protected from the interfering cosmic rays by 1200 m overlaying rock (3500 m.w.e). (ii) Gallex logo (iii) Gallex main building in hall A. The counting building is behind the main building, here barely visible. (iv) Counter shield tank (operated in a radon protected Faraday cage). Up to 24 counters can be accommodated simultaneously. See <sup>16</sup> for details. (v) extraction scheme: tank - absorber column – lab-scale reduction - chloride-hydride conversion (vi) (center): miniaturized (RSuprasil quartz gas proportional counter, active volume:  $\approx 1 \text{ cm}^3$ ).

The Gallex/GNO project lasted from 1983-2005, solar neutrinos were recorded between 14.05.1991 and 09.04.2003. Since 1998, this includes GNO (Gallium Neutrino Observatory), when Gallex was planned to be transformed in a permanently operating astronomical solar neutrino observatory. Unfortunately, these plans were terminated earlier than intended for external reasons that were unrelated to the operation of Gallex/GNO.

After the first year of full-scale operation 15 measurements ('runs') of the production rate of <sup>71</sup>Ge were carried out and the Gallex I result was submitted by the Gallex Collaboration to Physics Letters B on 31 May 1992<sup>16</sup>, back to back the same day with the interpretative Gallex Collaboration paper<sup>19</sup>. Simultaneously, on 8 June 1992 this 'historical first observation of ppneutrinos' was announced at the Neutrino 92 Conference in the Alhambra gardens of Granada. The result,  $83 \pm 21$  SNU (1 $\sigma$ ), implied a definite contribution from pp-neutrinos and thus, their discovery (Figure 11). 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' (Figure 12).



Figure 11 – Gallex announces first observation of solar pp-neutrinos at 'Neutrino 92' in Granada.

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Figure 12 – Neutrino 92 Conference Summary response by Alvara de Rujula.

In the foreword of the conference proceedings, Antony 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<sup>14</sup>.

With the Gallex I data in hand in 1992, Figure 13 displays the respective implications.

Physics Letters B285 (1992) 376 Citation index 31.5.92: #5+#11
Physics Letters B285 (1992) 390 14 RUNS
≈ 105 % of the pp- expectation
⇒ Hydrogen fusion in the solar interior experimentally observed
≈ 60 % of the total SSM- expectation
⇒ Definite deficit of pp- and/or <sup>7</sup>Beneutrinos observed

Figure 13 – Experimental situation after Gallex I in 1992.

The observed neutrino flux was (about 40 percent) short of what one must expect from the solar energy output (Solar constant). This reduction is highly suggestive of neutrino mass mediated oscillations between electron- muon- and tauon neutrinos on the way from the solar core to the detector on Earth. This is because the detection reaction, namely inverse beta decay on <sup>71</sup>Ga detects only electron neutrinos and is blind to the other neutrino flavors. However, within the  $2\sigma$ -range of the statistics at this time, the Gallex result was still compatible with the full SSM prediction. In this sense, neutrinos that have come through unaltered were still allowed <sup>19</sup>.

Ironically, the positive observation of pp-neutrinos (an ever-lasting cornerstone of modern astronomy) became overshadowed by non-objective wishful thinking of many to prematurely interpret the deficit relative to the SSM expectation value  $(83\pm21)$  SNU vs.  $(127\pm20)$  SNU as matter of fact indication of neutrino mass. A neutrino shortage from either pp- or <sup>7</sup>Be- neutrinos was evidently indicated, but the significance was  $2\sigma$  only. The Gallex collaboration discussed the issue in depth<sup>19</sup> and refrained from a respective claim in the professional spirit that claims of seminal importance must be  $>3\sigma$  at least <sup>14</sup>. Of course, it is nevertheless legitimate and state of art to discuss quantitatively the consequences if the result (central value) would persist while errors will shrink. Condition to this, the respective exercise is obviously contained in the Gallex interpretational paper<sup>19</sup>. It concludes that in a MSW<sup>c</sup> controlled neutrino flavor mixing scenario this would point to a neutrino mass of order  $3 \text{ meV/c}^2$  at small ( $\sin^2 2\theta$  of order  $10^{-2}$ ) or at large  $(\sin^2 2\theta \approx 0.6)$  mixing angles. In the following years Gallex worked hard to replace unsupported premature claims of others by improving statistics through adding more and more runs<sup>21,22</sup> and systematics by experimentally demonstrating the reliability of the radiochemical method by means of controlled calibrations with strong low energy neutrino sources (using  ${}^{51}Cr {}^{21,23}$ ) and chemical extraction yield control by spiking experiments (using <sup>71</sup>As <sup>24</sup>).

The Mega-Curie(!) chromium source systematic performance check  $(1^{st} \text{ priority})$  and statistically accurate calibration  $(2^{nd} \text{ priority})$  was a large-scale unique experiment of its own, using totally new technology. It involved the production of record quantities of isotopically enriched  ${}^{50}$ Cr (40 kg Cr enriched to 38.6%  ${}^{50}$ Cr), and record neutron doses for activation of  ${}^{50}$ Cr to  ${}^{51}$ Cr under rigorously enforced purity conditions. A team of CEA-Saclay, masterminded by Michel Cribier and Michel Spiro, took responsibility to neutron activate this enriched chromium at the Siloé reactor in Grenoble and to produce a Mega Curie  ${}^{51}$ Cr-neutrino calibration source. Figure 14 diplays the scheme of the source neutrino exposure in the identical setup in which the solar neutrinos were measured. Two  ${}^{51}$ Cr neutrino source experiments were performed in 1994 and 1995 and verified the proper operation of Gallex. The overall result is that it is factually demonstrated that there are no surprising bugs in the Gallex setup, the data produced are trustworthy.



Figure 14 – Configuration scheme of the GALLEX  $^{51}$ Cr neutrino source experiment.

<sup>&</sup>lt;sup>c</sup>Mikheyev-Smirnov-Wolfenstein' matter oscillations <sup>20</sup>

The significance for the mass mediated electron neutrino disappearance developed with improving statistics and allowed us to claim non-zero neutrino mass based on documented facts in 1995/96, with the overall result of  $(70 \pm 8)$  SNU from altogether 53 runs (end of Gallex III<sup>14</sup>. After Gallex IV, the end of Gallex data taking in 1997 (65 runs, left part of Figure 15) the result was 78 ± 8 SNU<sup>15</sup>. This was more than  $6\sigma$  below the SSM prediction. It was even significantly below a hypothetical minimal value that one obtains by simply adding the pp-flux requested from the solar luminosity and a <sup>7</sup>Be- $\nu$  flux not higher than what is needed to account for the <sup>8</sup>B-neutrino flux (via the <sup>7</sup>Be(p, $\gamma$ )<sup>8</sup>B reaction in the Sun) that was found in the Homestake and Kamiokande experiments at that time. Hence, in 1997, disappearance of bulk (sub-MeV) neutrinos was finally assured at >99% confidence level.

On top of this, in addition to the Cr-source experiment, another rigorous test was performed in 1997. Freshly produced <sup>71</sup>As was added to the gallium chloride target solution where it produces, by a weak decay process (mimicking neutrino interaction) <sup>71</sup>Ge, just as solar neutrinos do, and its recovery is recorded. Here one finally gets the statistics that is never attainable with solar neutrinos. Altogether, we used  $\approx 30~000~^{71}$ As atoms, what a signal! But: one could do this only after completion of the solar runs (with just a few events per month), since one breaks the purity seal. We did it after the end of Gallex and long before GNO runs started in 1998, where everything was re-conditioned anyhow. The result was perfect: 99+% recovery. This further 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 provided the first undisputed experimental evidence for 'New Physics'.

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 5/1998 to 4/2003. Altogether, Gallex and GNO have collected in 10 active years solar neutrino data in 123 runs. The single run results ('Davis-plot') are shown in Figure 15.



Figure 15 – Single run results for Gallex and GNO, altogether 123 runs  $^{25}$ .

Considering the additional active internal counter calibrations that were possible only after the end of their use in solar runs because of the contamination risk before, the final result for Gallex and GNO combined is  $67.6 \pm 5.1$  SNU<sup>18</sup>. In this context it is worth mentioning that the data show no obvious signs of any time dependence. We conclude with the satisfying notion that the early 'Granada' Gallex result of  $83 \pm 21$  SNU is fully compatible with the 12 years result of  $68 \pm 5$  SNU,  $(0.7 \sigma)$  from the now much more precise value<sup>25</sup>. <u>Credits</u>: Essential were both, great support of (European) funding institutions: MPG, BMFT, Krupp Foundation, INFN, CEA; and the enthusiastic support of the pioneers that got the LNGS underground laboratory going: Antonino Zichichi, Nicola Cabibbo, Luciano Maiani, and Enrico Bellotti. The latter was the first director of LNGS, and - at the same time – member and great supporter of Gallex in the critical initial phase, when LNGS was still in *statu nascendi*. More atmospheric information about the smooth functioning within the collaboration can be found in  $1^5$ .

## 6 The SAGE Gallium Experiments

Parallel to Gallex, another gallium solar neutrino experiment was performed by the Soviet-American Sage collaboration in the Baksan Valley (Caucasus) underground laboratory <sup>26</sup>. The shielding depth is  $\approx 4700$  m.w.e. In this case, metallic gallium is used as a target, where the Ge/Ga separation occurs in a two-phase emulsion that is more problematic. Gallium is kept liquid above 30 centigrade. The separation of germanium from gallium is achieved by mixing the gallium with an aqueous solution that is acidic (HCl) and oxidizing (H<sub>2</sub>O<sub>2</sub>). This occurs in up to 8 Teflon-lined reaction vessels with provisions for easy stirring. Each vessel accommodates up to 6 tons of gallium. In order to separate germanium from gallium, a disperse emulsion is formed. 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 principal like those described above for Gallex.

An advantage claimed for the Sage process is to have potentially fewer side reactions because the target contains fewer free protons than the aqueous solution used in Gallex. On the other hand, there is always concern about potential withholding mechanisms in delicate experiments at the few-atoms level. This favors classical ion chemistry of aqueous solutions over surface chemistry in a heterogeneous two-phase emulsion. The latter process is theoretically much less understood: we deal with the kinetic behavior of (isotopically) identified single atoms.

The process is also more elaborate in practical aspects. Each extraction run involves handling of 8 vessels. Substantial quantities of new chemicals must be added in each run. This makes it difficult to ensure the radiochemical integrity of the target. These are the objective reasons why skepticism of the scientific community prevailed in evaluating Sage data releases before a convincing demonstration of systematical errors in such a completely new technology had been experimentally assured.

The Sage extractions have been done with variable amounts of gallium, up to 57 tons. Altogether, about 60 solar runs were done between 1990 and 1997 under somewhat variable conditions of <sup>71</sup>Ge-counting <sup>27,28</sup>. They started solar neutrino recording in January 1990 and communicated a nil result after 6 months, this was advertised as indication of New Physics, but the claim was never made in a professional form and it 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 6/1991-6/1992. This combined data set of 14 runs (retrospectively specified as 'Sage 1') was quoted as  $81 \pm 20$  SNU (1 $\sigma$ ), a few weeks after the practically identical Gallex I result was released in Granada and Physics Letters, see Figure 16 where the results of Gallex and Sage until 1998 are compared. Good agreement with the final result is established from 1993 onwards, but not before.

A successful <sup>51</sup>Cr neutrino source experiment was performed in 1996 by the Sage collaboration as well <sup>29</sup>. Less source strength (or target mass) was required to produce the same number of <sup>71</sup>Ge atoms because gallium metal is denser than gallium chloride solution. The Sage source had a strength of 517 ± 6 kCi and was used to irradiate 13.12 tons of gallium in a single reaction vessel. The experimental result expressed as cross section is  $\sigma = (55.2 \pm 6.6) \times 10^{-46} \text{ cm}^2 (1\sigma)$ . This is consistent with full response of the Sage detector.



Figure 16 – Published Gallex and Sage data for comparison.

# 7 Synopsis (as of the end of the past millennium)

The status of the major results from the pioneering solar neutrino experiments at the end of 1998 is summarized in Figure 17.



Figure 17 – Status of the major results from the pioneering solar neutrino experiments, end of 1998.

- All solar neutrino experiments observe a signal above zero.
- All solar neutrino experiments observe less neutrinos than predicted from stellar theory.
- The observed signals are  $\approx 1/3$  of expectation for the chlorine detector and  $\approx 1/2$  of expectation for the Kamiokande and Gallium detectors.

The confidence in the relevance of the SSM is high due to the good agreement of the solar density profile with the helio-seismological observations.

Initially, the full predicted value for the SSM was still within  $2\sigma$  of the Gallex I result. In the following years (>1992) the error shrank and first the solar <sup>7</sup>Be- $\nu$  deficit and afterwards the pp- $\nu$  deficit became significant. A plain reduction of the <sup>8</sup>B- $\nu$  was no longer sufficient to reproduce the evidence. Consequently, the emphasis shifted to particle physics as the most likely cause of the missing electron neutrinos. Neutrino flavor oscillations became the most favored option in spite (or just because!) of the fact that this would imply non-zero mass eigenstates, not included in the standard particle model.

Neutrino oscillations imply non-standard neutrinos, irrespective of particular theoretical models. The most natural approach is to assume a see-saw neutrino mass mechanism and to assign the dominant mixing of solar neutrinos to  $\nu_e \rightleftharpoons \nu_\mu$  within a regular lepton-quark mass hierarchy, with  $m(\nu_e) \ll m(\nu_\mu)$ . With the available experimental data in Figure 17, this led to a mass estimate for the muon neutrino of  $m(\nu_\mu) \approx 2.2$  meV for a small mixing angle of  $\theta = 2.2^{\circ}$  ('small angle solution'). Today, with available results from the SNO and Super-Kamiokande experiments, we know that vacuum oscillations dominate below 1 MeV and the 'large mixing angle' is  $\theta = 36^{\circ}$ . Without the pp-sensitive gallium experiment, this energy domain would not have been accessible (above  $\approx 1.5$  MeV matter oscillations controlled by the MSW<sup>d</sup> effect take over).

<u>Concluding remark</u>: Radiochemical solar neutrino experiments led to great path-making discoveries till the turn of the last millennium. This phase ceased with the advent of real-time experiments (Super-Kamiokande, SNO, Borexino, KamLAND) that now allow to observe multiple parameters synchronously, not just reaction rates only. These present developments would not exist without the pioneering precursors that are memorized in the present article.

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 $<sup>^{</sup>d}$ Mikheyev-Smirnov-Wolfenstein effect, considering the influence of dense matter on neutrino oscillations.

<sup>&</sup>lt;sup>e</sup>In preparing this article in the historical context, some segments from my own (single authored) publications <sup>10,13,14,15,17,30</sup> were partially re-used in a respectively adapted format for the present context.

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