

Radiochemical Solar Neutrino Experiments, “Successful and Otherwise”

Richard L. (Dick) Hahn

*Solar-Neutrino & Nuclear-Chemistry Group**

Chemistry Department, BNL

Upton NY 11973

***Research sponsored by the Offices of Nuclear Physics and High Energy Physics,
Office of Science, U.S. Department of Energy**

Neutrino 2008

Christchurch, NZ May 26, 2008

>40 Years of Neutrino R&D @ BNL Chemistry Dep't.

Nuclear and Radiochemistry

- **Done:** **HOMESTAKE** Radiochemical Detector



- **Done:** **GALLEX** Radiochemical Detector



- **Done:** **SNO** Water Čerenkov Real-time Detector

Ultra-pure D_2O (1996 - \geq 2006)

- **New:** **#1 Focus - THETA-13**

High-Precision Experiments at [Daya Bay Nuclear Reactors](#)

Real-time Detector (R&D) Gd in [Liquid Scintillator](#), [Gd-LS](#) (began 2004)

- **New:** **SNO+** Real-time Detector (R&D) at **SNOLab**

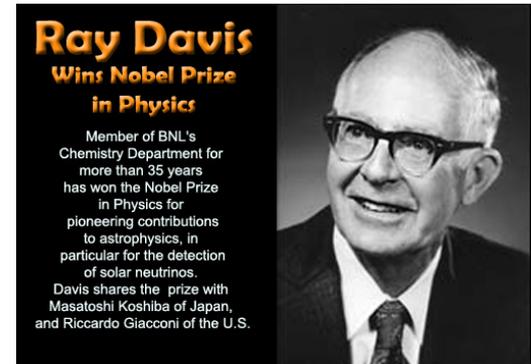
- [\${}^{150}\text{Nd-LS}\$](#) (began 2005) **Double beta-decay**

- **New:** **LENS, MiniLENS** Real-time Detector (R&D)

- [\${}^{115}\text{In-LS}\$](#) (began 2000), Detect pp and ${}^7\text{Be}$ Solar Neutrinos

- **New:** **Very Long-Baseline Neutrino Oscillations**

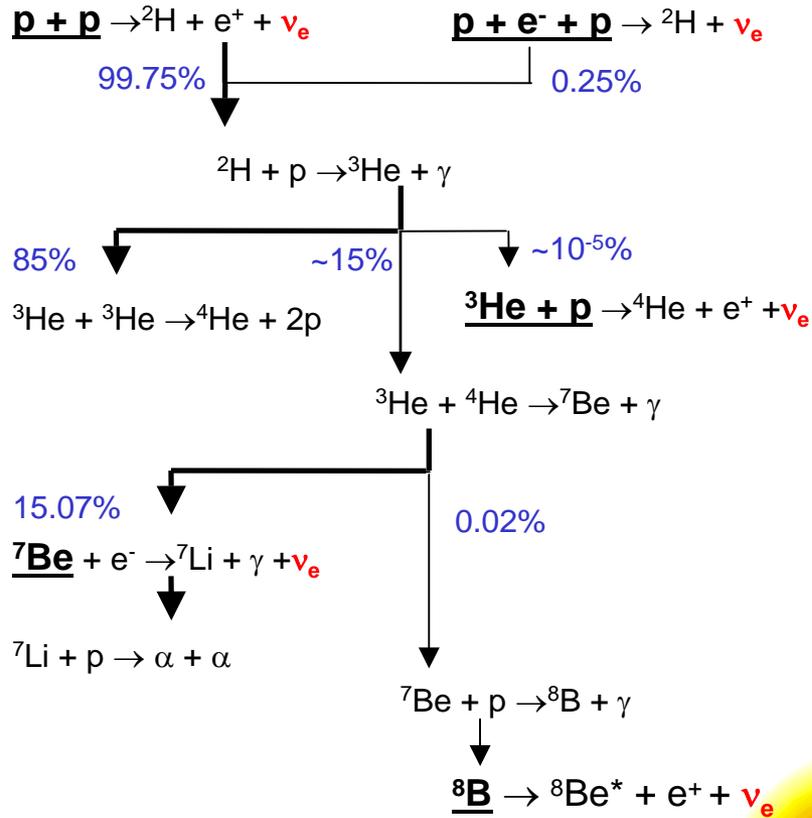
- ν_μ Beam from Accelerator to DUSEL (R&D began 2002)



Note: Hahn became Leader of BNL Group in February 1987, the same time as SN-1987A

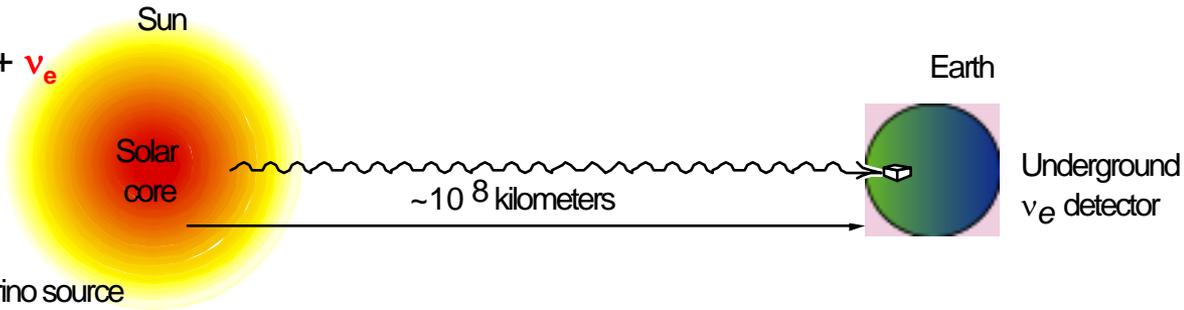
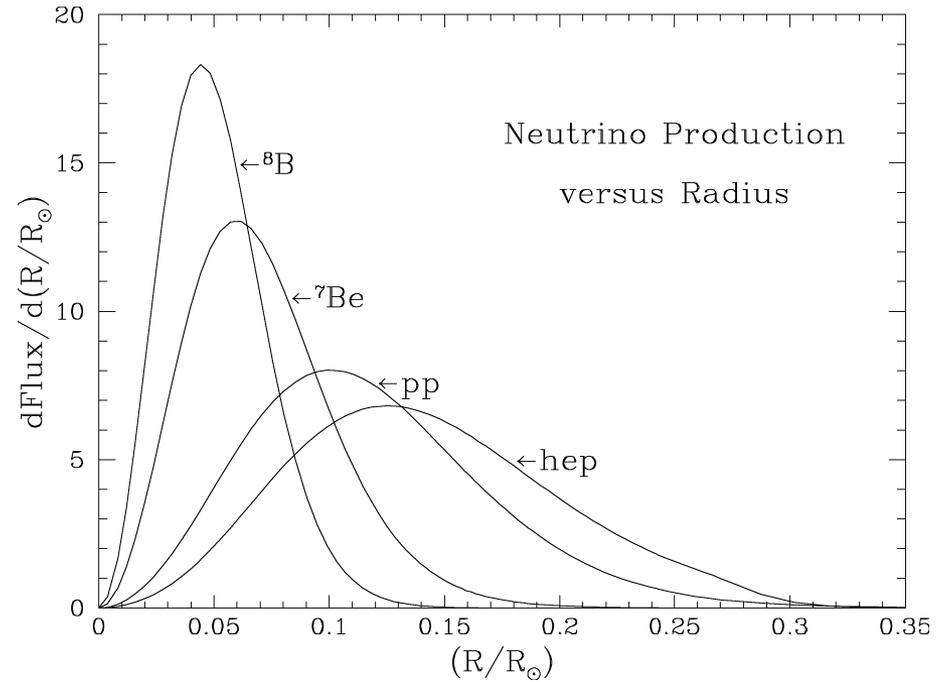
Neutrino Production in the Sun

Light Element Nuclear Fusion Reactions



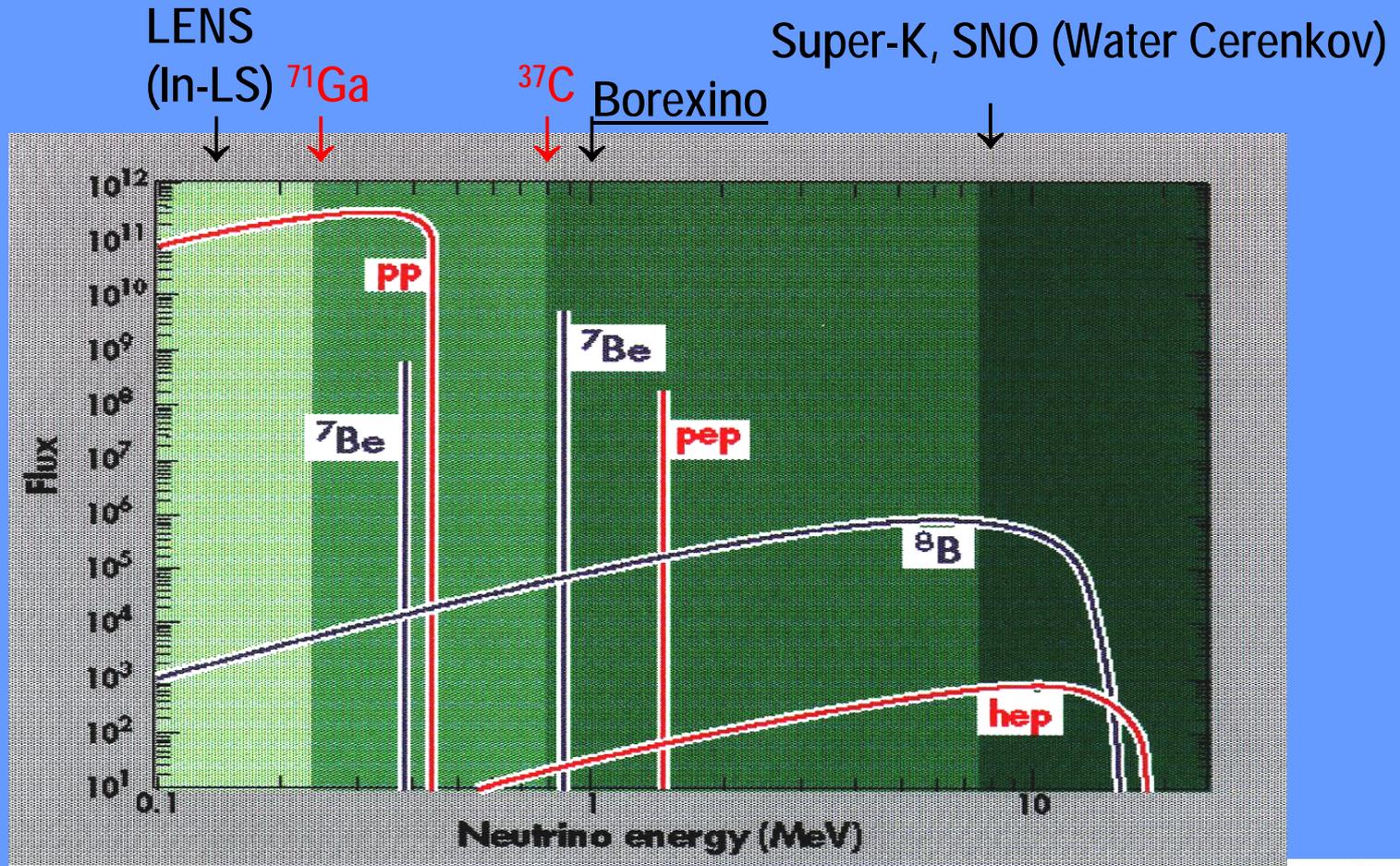
Primary neutrino source
 $p + p \rightarrow \text{D} + e^+ + \nu_e$

Neutrino Production Radius



Predicted Energy Spectra of Solar Neutrinos from the Standard Solar Model (SSM – Bahcall et al.)

Arrows ↓
Denote
Experimental
Thresholds



Radiochemical Solar Neutrino Detectors ⁺

⁺ John Bahcall, Neutrino Astrophysics, Cambridge U. Press, 1989, chap. 13, p. 363

Nu Capture, $\nu_e + (A, Z) \rightarrow e^- + (A, Z+1)^*$

✓ $^{37}\text{Cl} \rightarrow ^{37}\text{Ar}$ ($T_{1/2} = 35.0$ d, E-threshold = 0.814 MeV)

✓ $^{71}\text{Ga} \rightarrow ^{71}\text{Ge}$ ($T_{1/2} = 11.4$ d, E-threshold = 0.233 MeV)

X $^{127}\text{I} \rightarrow ^{127}\text{Xe}$ ($T_{1/2} = 36$ d, E-threshold = 0.789 MeV)

? $^7\text{Li} \rightarrow ^7\text{Be}$ ($T_{1/2} = 53$ d, E-threshold = 0.862 MeV)

? $^{81}\text{Br} \rightarrow ^{81}\text{Kr}$ ($T_{1/2} = 2 \times 10^5$ yr, E-threshold = 0.470 MeV)

(Geochemical – test concept of steady-state sun)

X $^{98}\text{Mo} \rightarrow ^{98}\text{Tc}$ ($T_{1/2} = 4 \times 10^6$ yr, E-threshold >1.74 MeV)

X $^{205}\text{Tl} \rightarrow ^{205}\text{Pb}$ ($T_{1/2} = 14 \times 10^6$ yr, E-threshold = 0.054 MeV)

Legend – “Successful and Otherwise”:

✓ = “Successful”, X = “Not successful”, ? = Did not get beyond R&D stage

Principles of Radiochemical Solar ν Detection



- Huge multi-ton detectors
- Locate deep underground; (p,n) reaction mimics ν capture
- Do in batch mode, with solar exposures $\sim 2 T_{1/2}$
- Do sensitive radiochemical separations to separate product chemical element (Z+1) from target Z: **isolate ~ 10 product atoms from $\sim 10^{30}$ target atoms.**
- Purify product, convert to suitable chemical form for high-efficiency, low-background nuclear counting
- Measured energy spectrum and half-life identify $(A, Z+1)^*$
- Note: $T_{1/2} \rightarrow \log\text{-ft value} \rightarrow \nu$ cross section (g.s. \rightarrow g.s.)

Concerning low-background nuclear counting

- **Einstein famously said that “God does not play dice.”**
- **A new aphorism (RLH): “The existence of Radon proves that God does play practical jokes.”**

Acknowledgements of Discussions and Slides for this Presentation

Ken Lande – ^{37}Cl , ^{127}I

Bruce Cleveland – ^{37}Cl , ^{71}Ga (SAGE)

Till Kirsten – ^{71}Ga (GALLEX, GNO)

Wolfgang Hampel – ^{71}Ga (GALLEX, GNO)

Vladimir Gavrin – ^{71}Ga (SAGE), ^7Li

(Kurt Wolfsberg – ^{98}Mo)

Not All ν Experiments have worked:

“Unsuccessful” Experiments - I

$^{127}\text{I} \rightarrow ^{127}\text{Xe}$ ($T_{1/2} = 36$ d, E-threshold = 0.789 MeV)

- o Developed by K. Lande et al. at U Penn to check the well-known Cl deficit
- o Chemistry used was analogous to the Cl experiment
- o Novel automated chemistry developed to segregate the product Xe into day and night fractions
- o Prototype testing was ended when Homestake Mine was shut down after the Barrick Co. purchased the mine and the water pumps were shut down

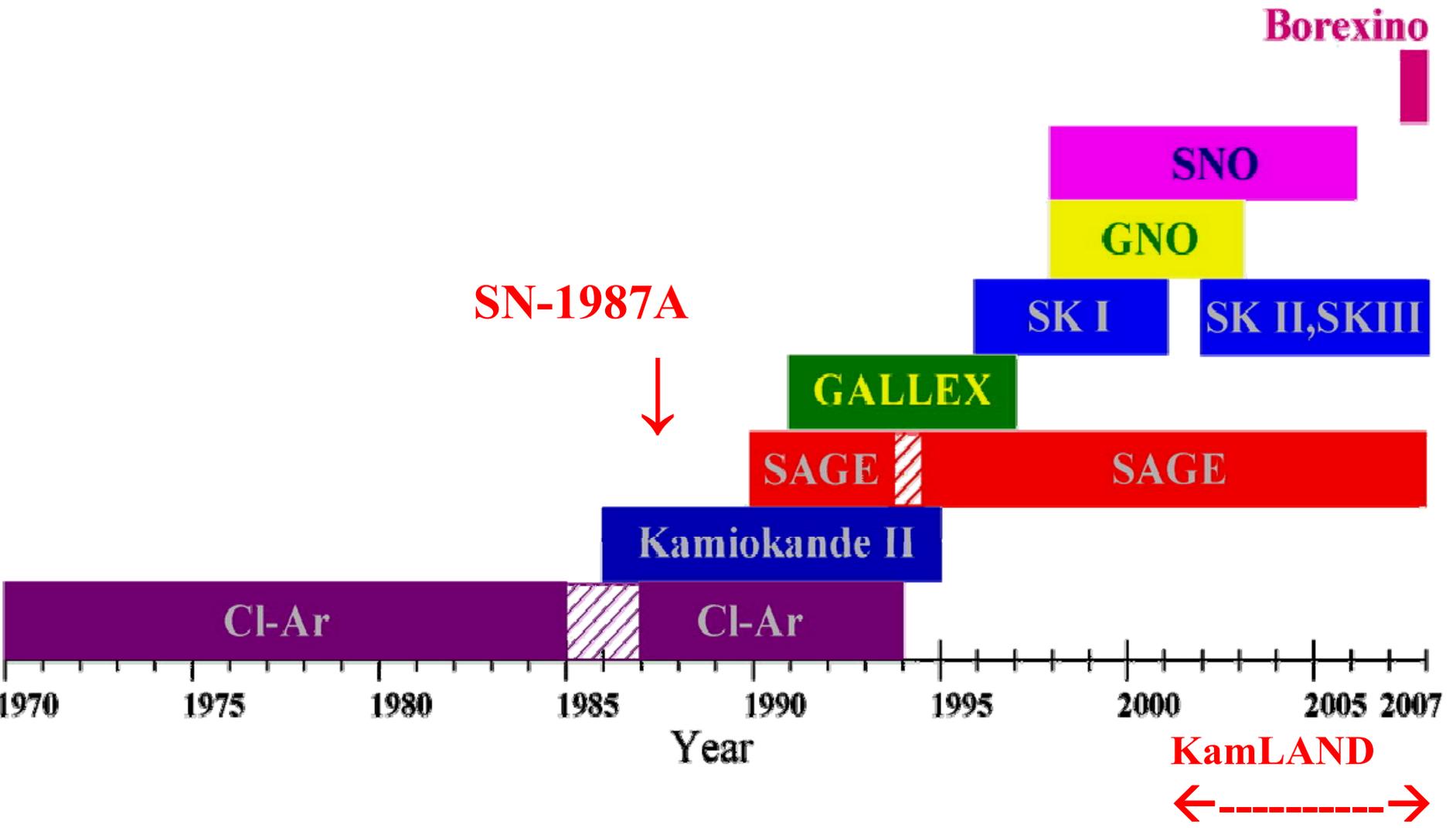
“Unsuccessful” Experiments - II



- **Developed by K. Wolfsberg et al. at Los Alamos to check the CI result over geological time scale**
- **At the Henderson Mine in Colorado, identified a deep Mo-sulfide ore body that was adequately shielded from cosmic rays**
- **Novel chemical system was developed to separate Tc from Mo; was installed at chemical smelter where MoS was roasted to MoO**
- **Actual experiment was run with the deep ore; however, the smelter equipment was contaminated with cosmogenic Tc from previously processed Mo from shallow ore deposits**
- **Result: Their experimental neutrino production rate was several times larger than the SSM value**
- **Funding was never obtained to redo the experiment with properly cleaned equipment at the smelter → so experiment was aborted**

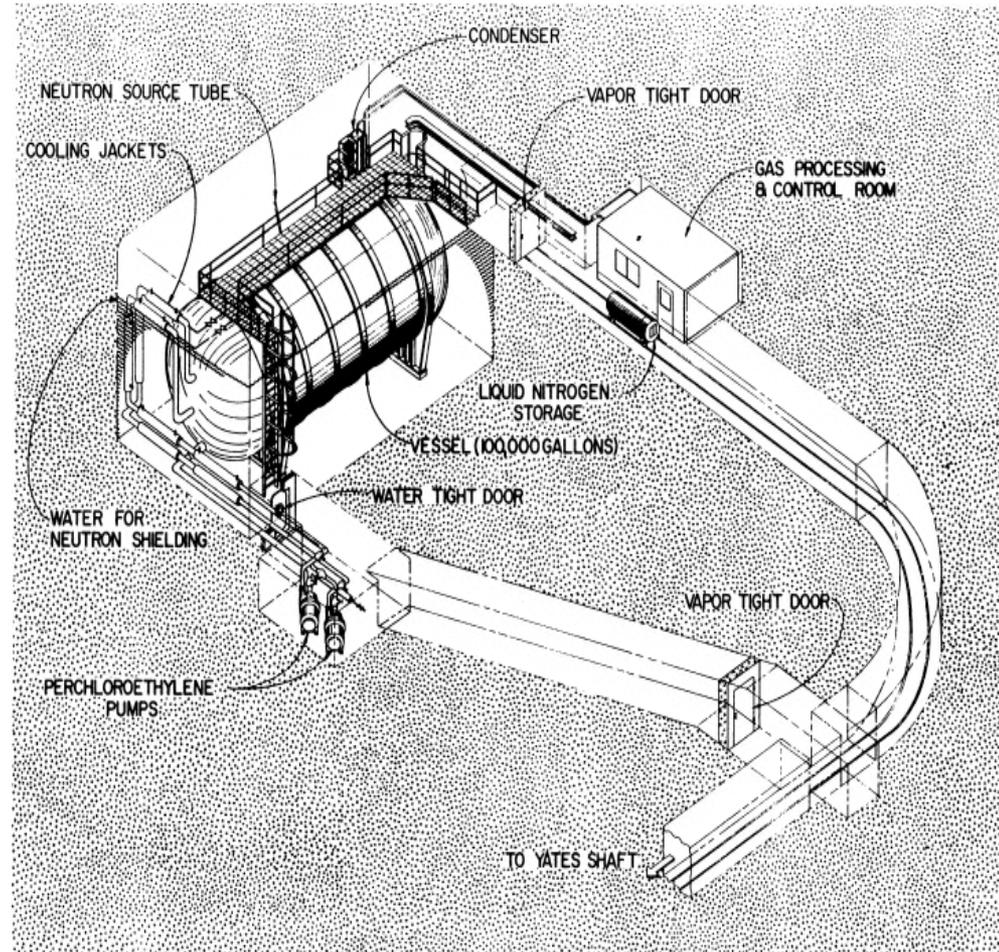
Time Line for Successful Solar ν Experiments

(by V. Gavrin, **modified by RLH**)

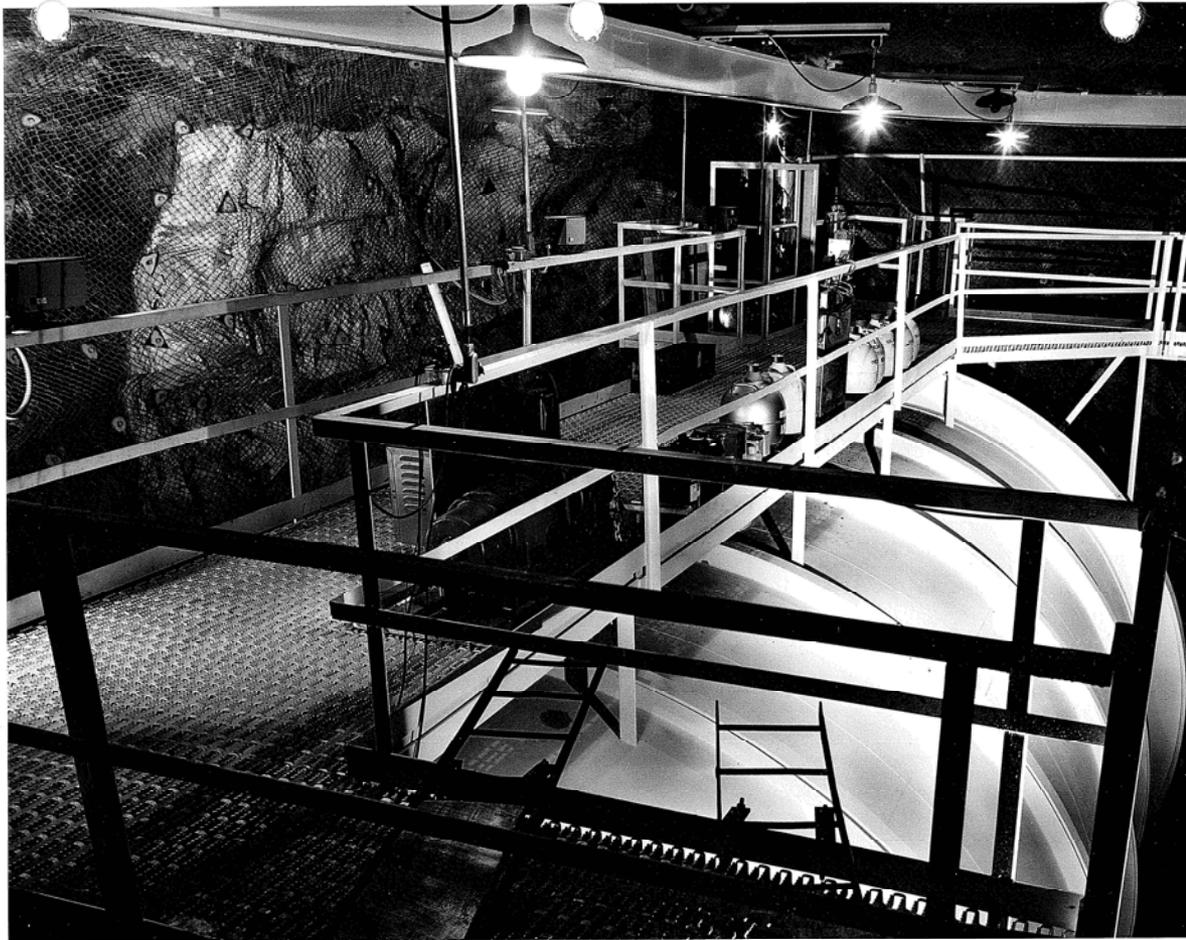


Birth of Solar Neutrino Experiments

- 1965-67: Davis builds 615 ton chlorine (C_2Cl_4) detector
- Deep underground to suppress cosmic ray backgrounds.
 - Homestake Mine (4800 mwe depth)
- Low background proportional detector for ^{37}Ar decay.
 - $^{37}Cl + \nu_e \rightarrow ^{37}Ar + e^-$
 - Detect $^{37}Ar + e^- \rightarrow ^{37}Cl + \nu_e$
($t_{1/2} \sim 37$ d)

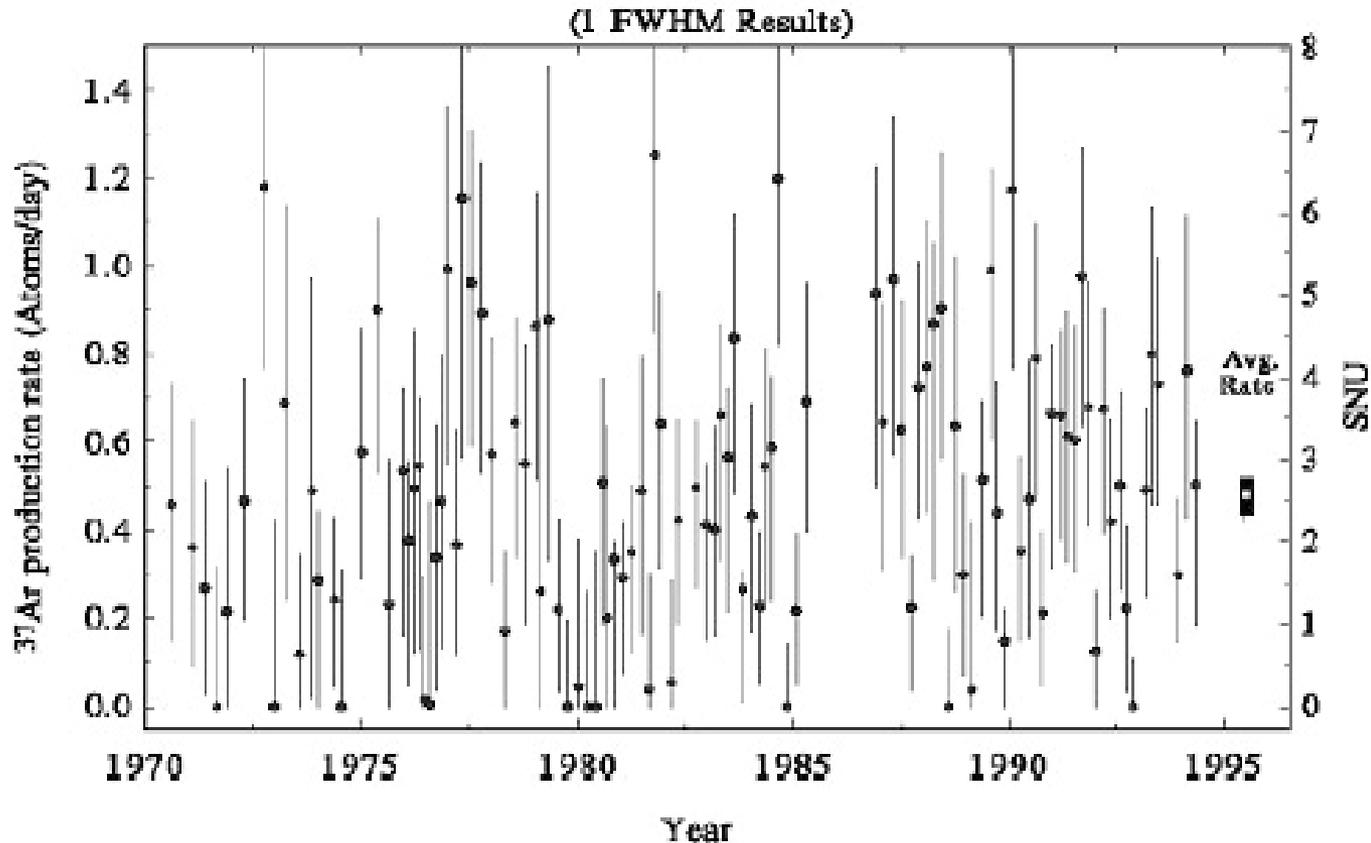


1965-Ray Davis began construction of the Cl Detector to look for ν_e from $H \rightarrow He$ fusion in the Sun



Chlorine Data 1970-1994

(Experiment was ended when Homestake Mine was shut down after the Barrick Co. purchased the mine)



**“Davis Plot”
with 108 runs,
reveals ν deficit**

Fig. 2 Final results of Davis experiment (Cleveland et al. 1998). The average rate of about 2.5 SNU is much lower than the calculated rate of about 8.6.

1 SNU = 1 neutrino capture per sec per 10^{36} target atoms

The Importance of Ray Davis's Discoveries

- He was the first to observe neutrinos from the Sun; he used a radiochemical method for detection.
- This result confirmed our theories that stars produce energy by nuclear fusion → 2002 Nobel Prize in Physics.
- But we scientists expected that result.
- More exciting for us, he observed an unexpected result, too few neutrinos compared to the SSM.
- This deficit became known as the Solar Neutrino Problem (SNP).
- Initially many doubters thought that Ray was wrong, but follow-on exp'ts all confirmed the ν deficit.

Personal Recollections About Ray

- After I rejoined BNL in 1987 to work on ν 's, Ray and I would speak often about neutrino experiments. He never quite understood how I could manage to operate in a collaboration as large as GALLEX, and the even larger size of SNO was almost incomprehensible to him
- In his later years, he worked in the lab at BNL, trying without success to develop a radiochemical method to detect geo-antineutrinos, using his existing Cl detector and the reaction $^{35}\text{Cl} + \text{anti-}\nu \rightarrow ^{35}\text{S}^* + e^+$
- Many times I commented to him that most younger scientists did not know that he had spent most of his career in the BNL Chemistry Dep't. I encouraged him to correct the misconception that he was a physicist. His reply: "Dick, I wasn't even a chemist when I was doing the Cl experiment, I was a plumber."

SAGE $^{71}\text{Ga} + \nu_e \rightarrow ^{71}\text{Ge} + e^-$

From Gavrin, TAUP-07

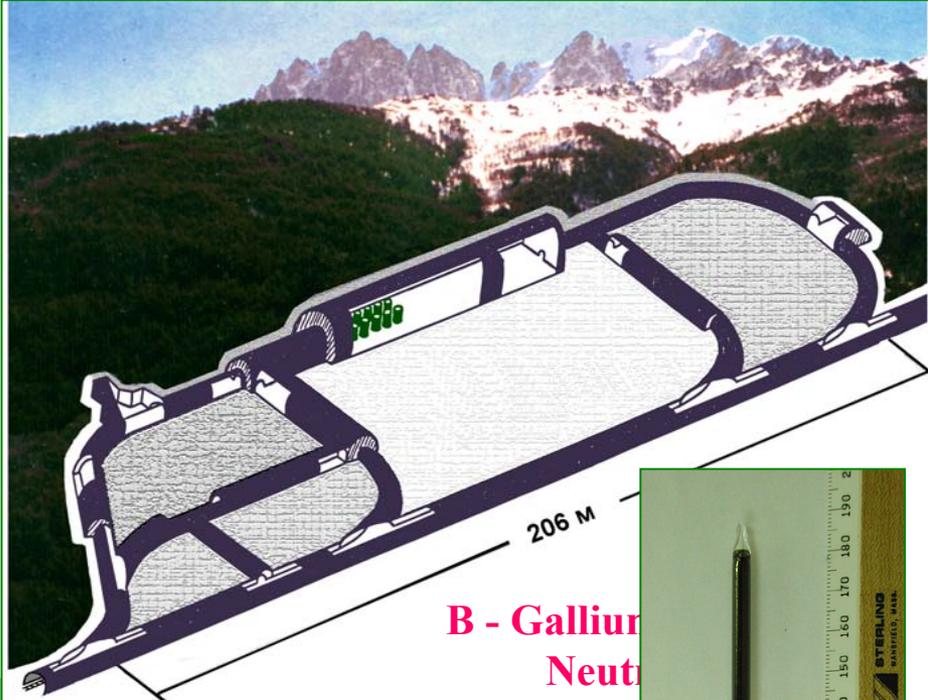
Baksan Neutrino Observatory, northern Caucasus,
3.5 km from entrance of horizontal adit,

2100 m depth (4700 m.w.e.)

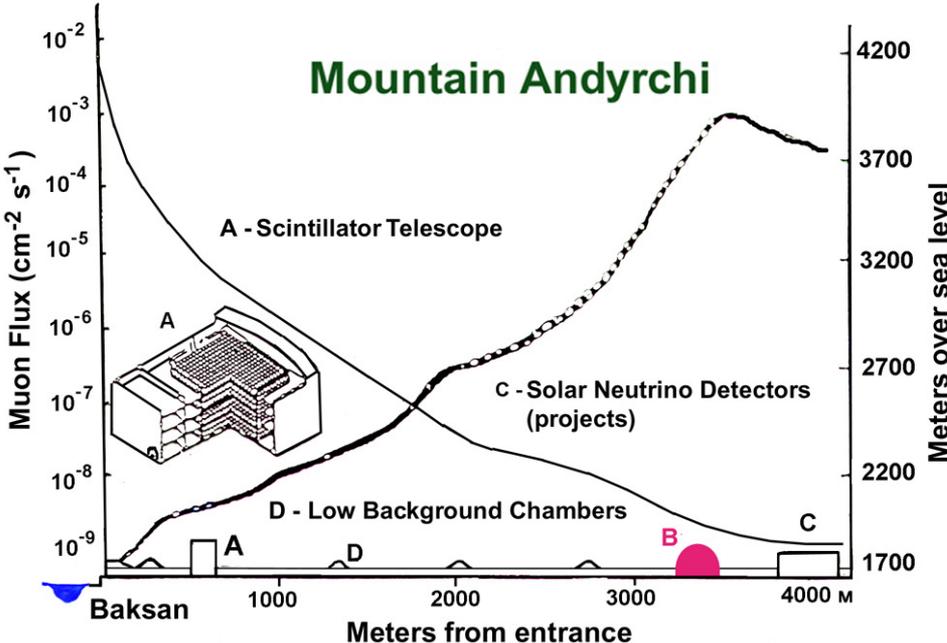
Data taking: Jan 1990 - till present,
50 tons of metallic Ga.

Atoms of ^{71}Ge chemical are extracted and
its decay is counted.

Sensitivity: One ^{71}Ge atom from $5 \cdot 10^{29}$ atoms Ga
with efficiency $\sim 90\%$



B - Gallium
Neutrino



SAGE

Measurement of the solar neutrino capture rate with gallium metal.

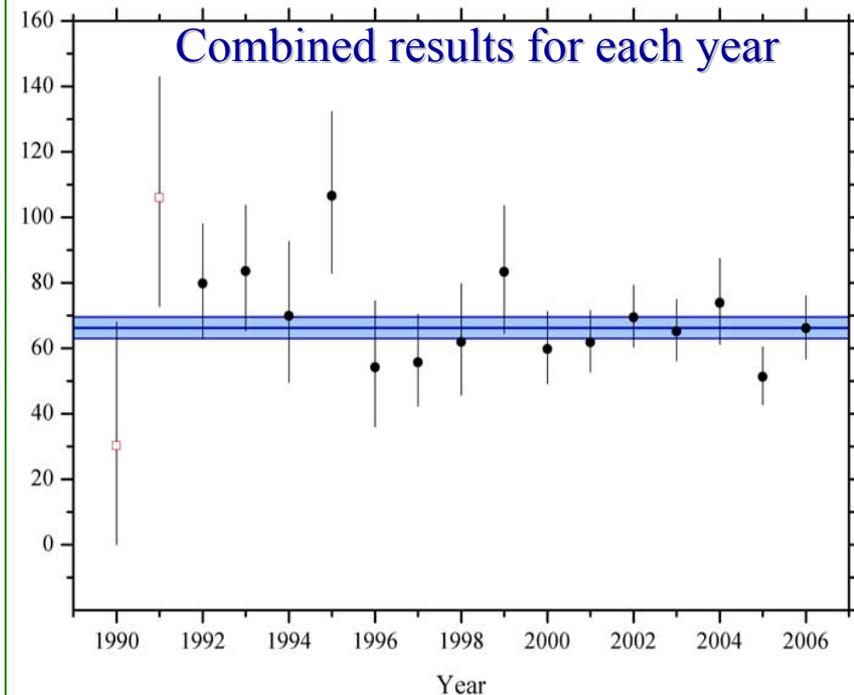
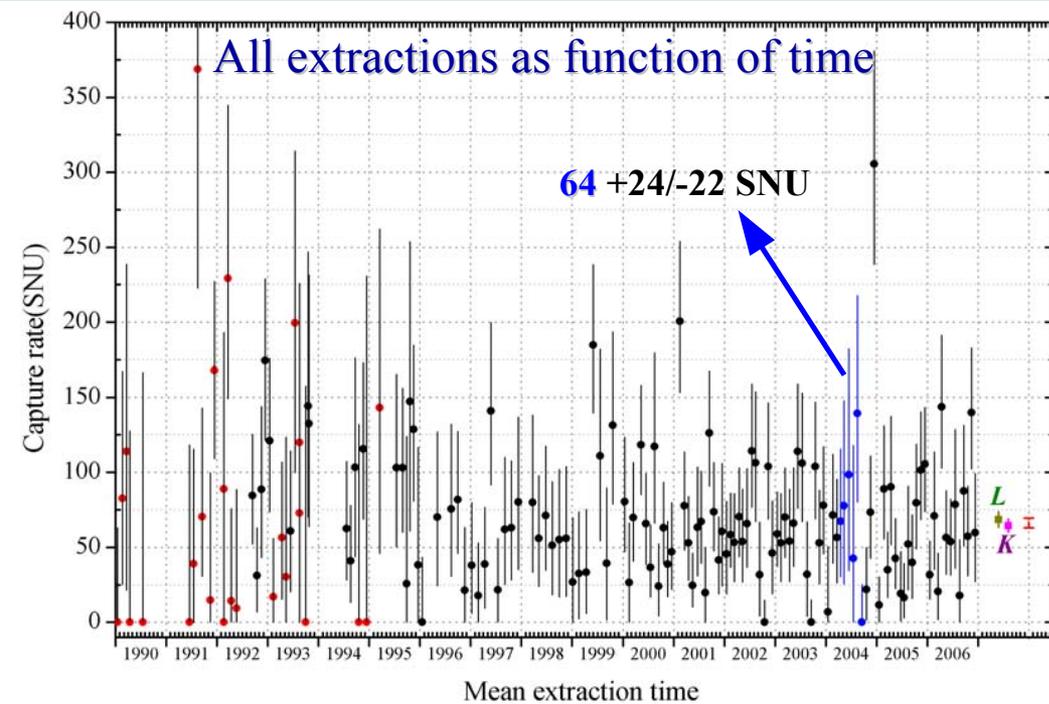


Presently **SAGE** is the **only** experiment sensitive to the *pp* neutrinos

It is one of the **longest almost uninterrupted time of measurements** among solar neutrino experiments

17 year period (1990 – 2006): 157 runs, 288 separate counting sets

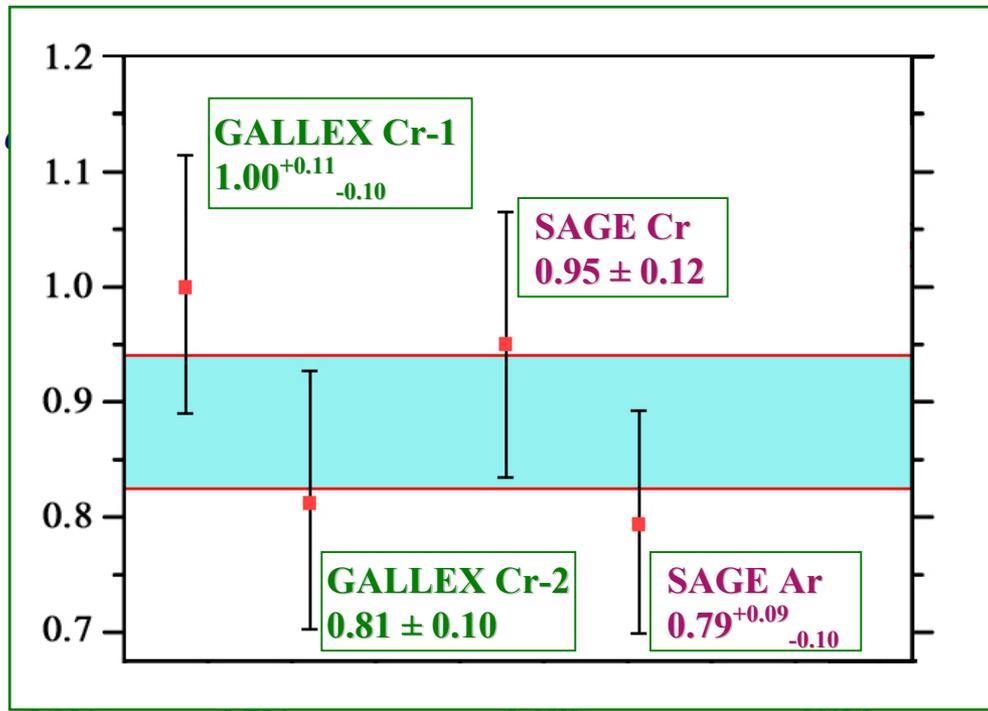
Results: $66.2^{+3.3}_{-3.2}$ $^{+3.5}_{-3.2}$ SNU or $66.2^{+4.8}_{-4.5}$ SNU (GALLEX 67.6 SNU)



SAGE continues to perform regular solar neutrino extractions every four weeks with ~50 t of Ga

Gallium

- m_{Ga} (tons)
- $m_{of\ target}$ (kg)
- enrichment (% ^{50}Cr)
- source specific activity (KCi/g)
- source activity (MCi)
- expected rate



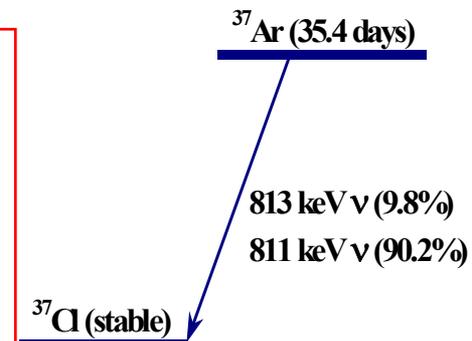
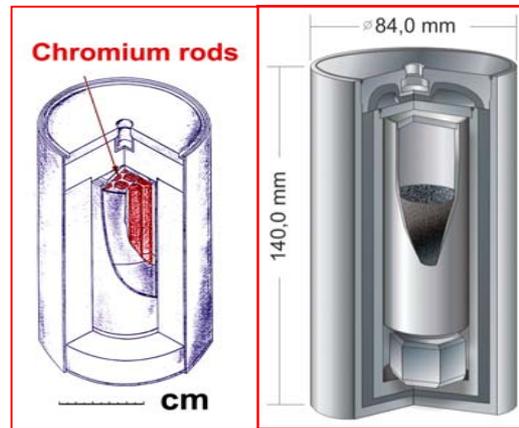
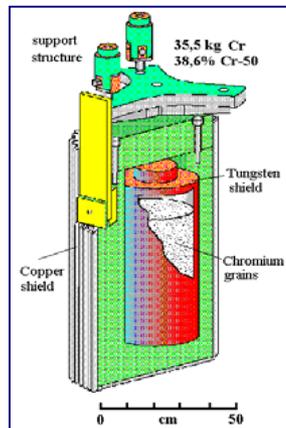
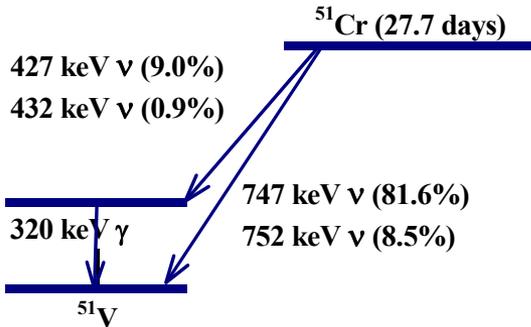
$R = p_{measured} / p_{predicted}$

1.0 ± 0.11 0.81 ± 0.10 0.95 ± 0.12 0.79 ± 0.1

$R_{combined}$

0.90 ± 0.07

0.86 ± 0.08

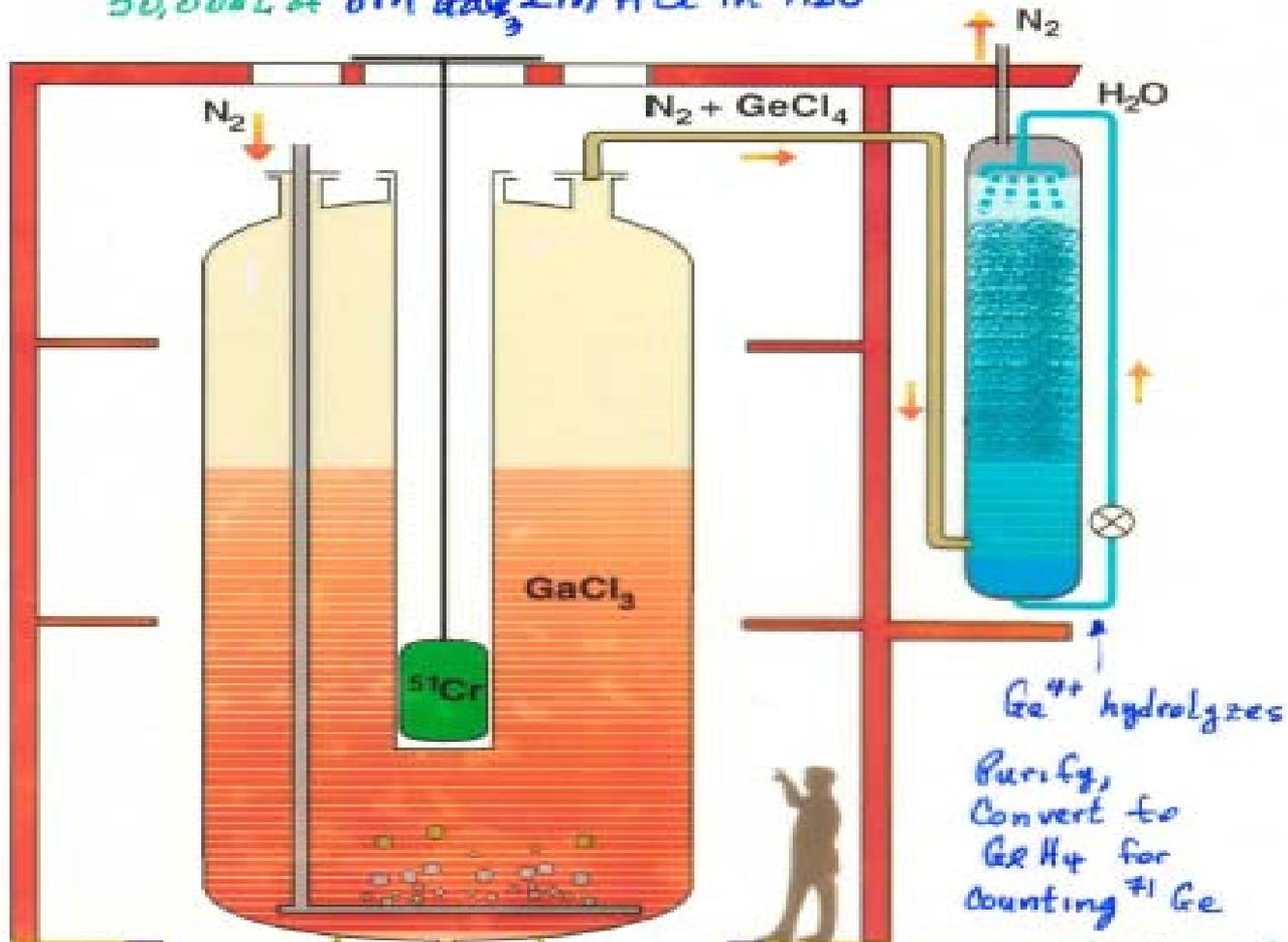


Future Plans for SAGE

- *Further solar running for the next three years. Want to get more pp data while Borexino is measuring ${}^7\text{Be}$ neutrinos.*
- *Measurement of the response of their Ga solar neutrino experiment to neutrinos from a ${}^{51}\text{Cr}$ source with accuracy better than 5% (>2 MCi).*
- *Are considering feasibility of using energetic protons at an accelerator to measure $B(\text{GT})$ for ${}^{71}\text{Ga} \rightarrow {}^{71}\text{Ge}$.*

G
A
L
L
E
X
a
t
L
N
G
S

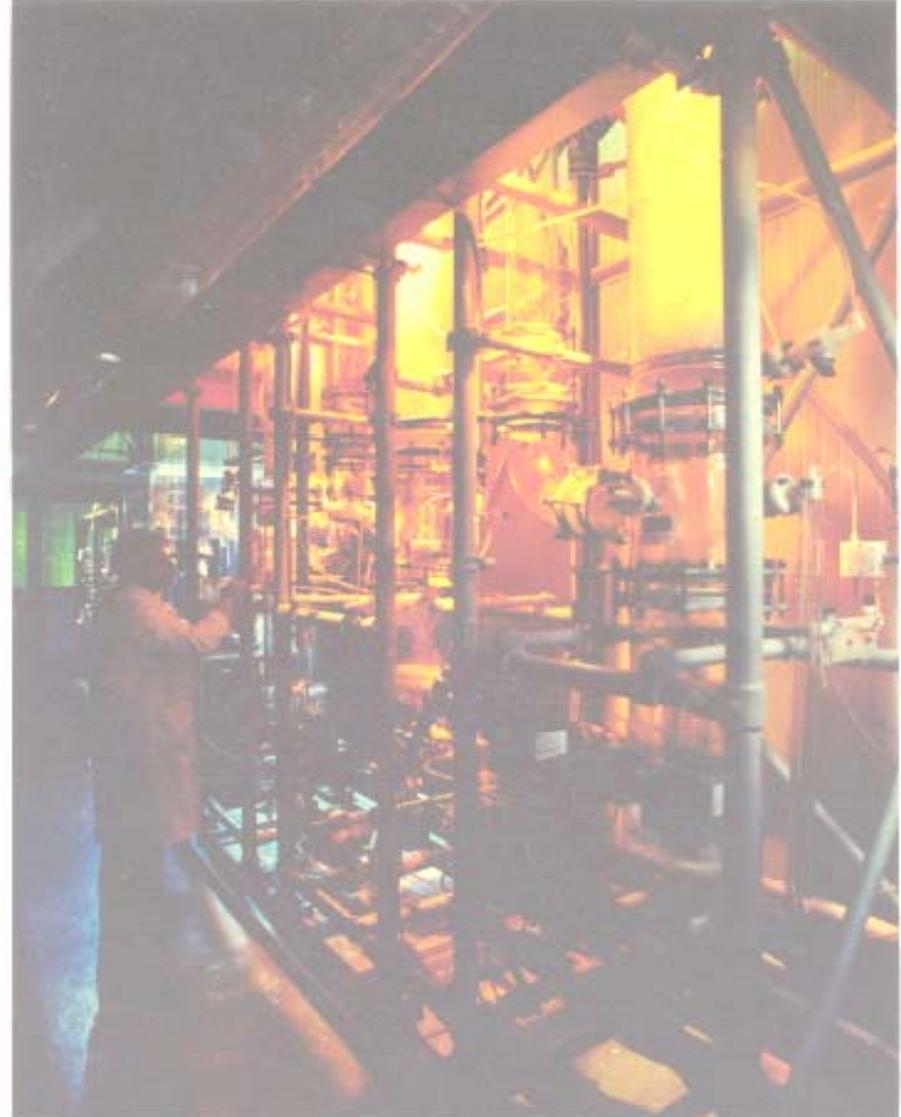
GALLEX : $^{71}\text{Ga} + \gamma \rightarrow ^{71}\text{Ge}^* + e^-$
THRESHOLD = 0.233 MeV
30.3 tons Ga in 100 tons of solution
50,000L of 8M GaCl_3 , 2M HCl in H_2O



Ge^{4+} hydrolyzes
Purify,
Convert to
 GeH_4 for
counting ^{71}Ge
 $\sim 10^{29}$ atoms Ga
Isolate ~ 10 atoms
Ge with $\sim 95\%$
efficiency.

R
A
D
I
O
C
H
E
M
I
C
A
L

GALLEX (65 runs - 1991-97) & GNO (58 runs - 1998-2003)



Recent Update from GALLEX*

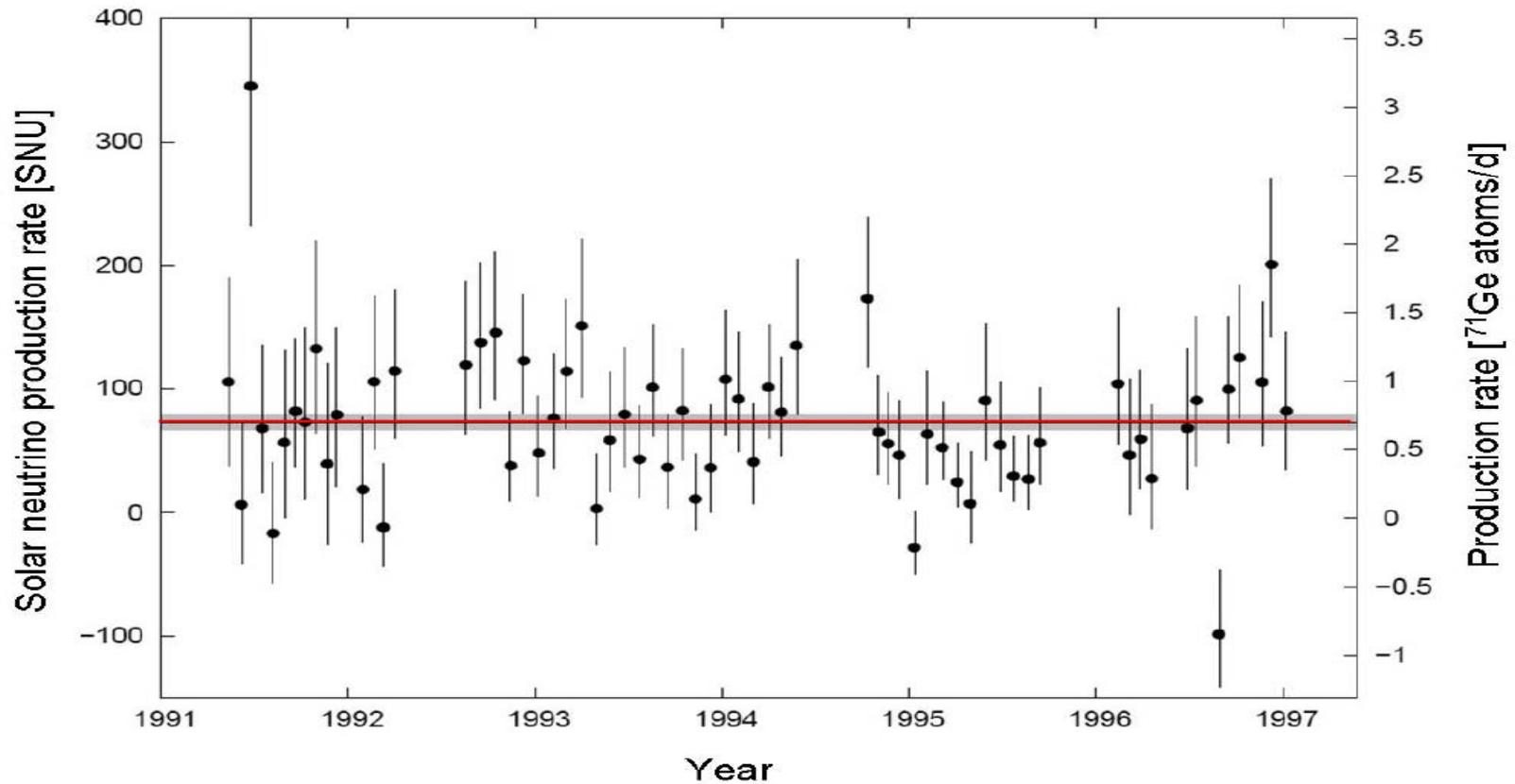
Results of a recent complete re-analysis of the GALLEX data:

- ❑ Recalibrated each counter with $\sim 10^5$ inserted Ge decays, which they could not have done before completing the low rate solar runs
- ❑ Did full-blown Pulse Shape Analysis (PSA) instead of the previously used Rise Time Analysis (RTA)
- ❑ Improved the Rn-cut efficiency and the Background determinations
- ❑ Also applied this new information to their Cr-source data

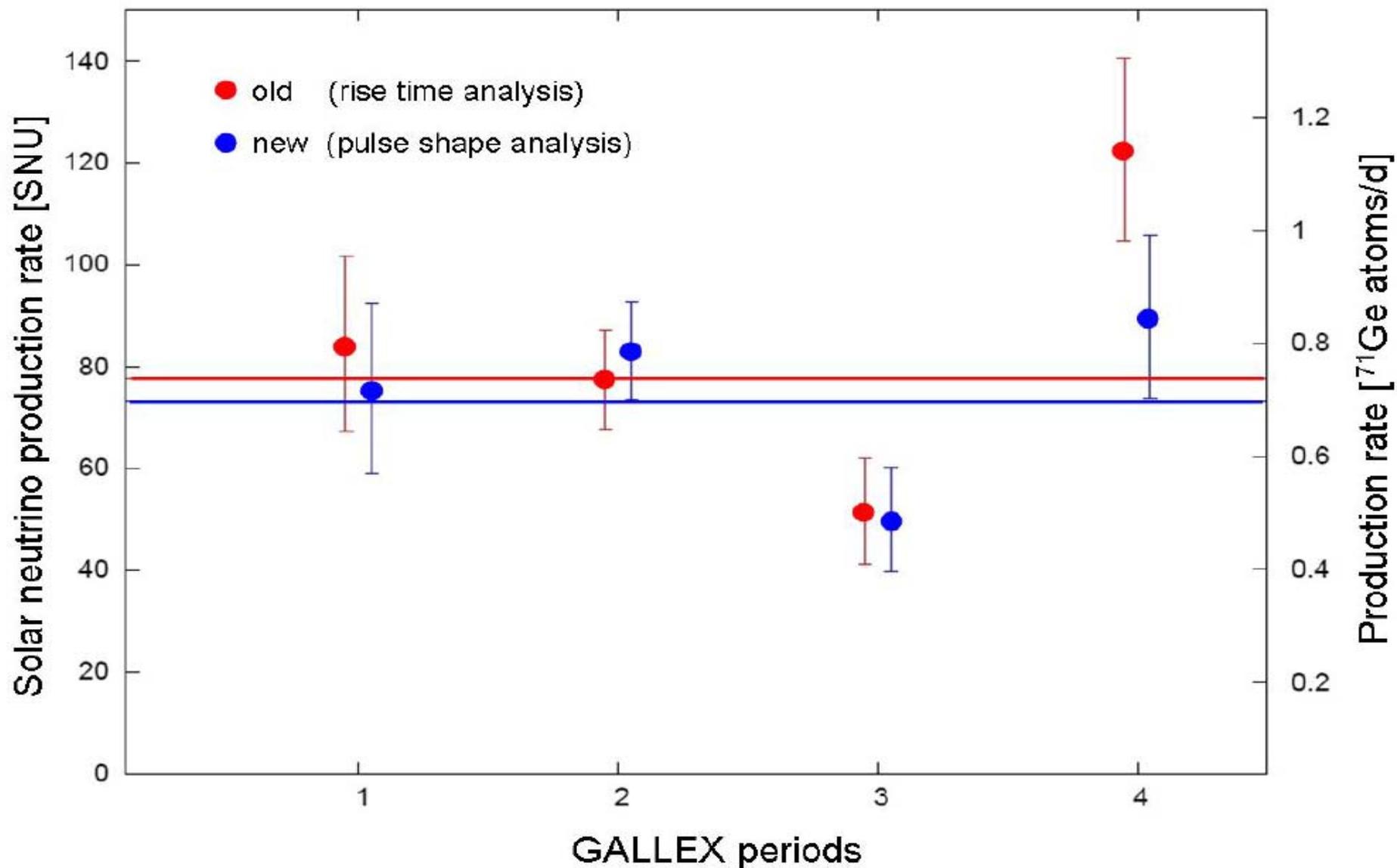
* From T.Kirsten TAUP-07; W. Hampel, T. Kirsten F. Kaether, May 2008

Updated Davis Plot from GALLEX

(May 08)

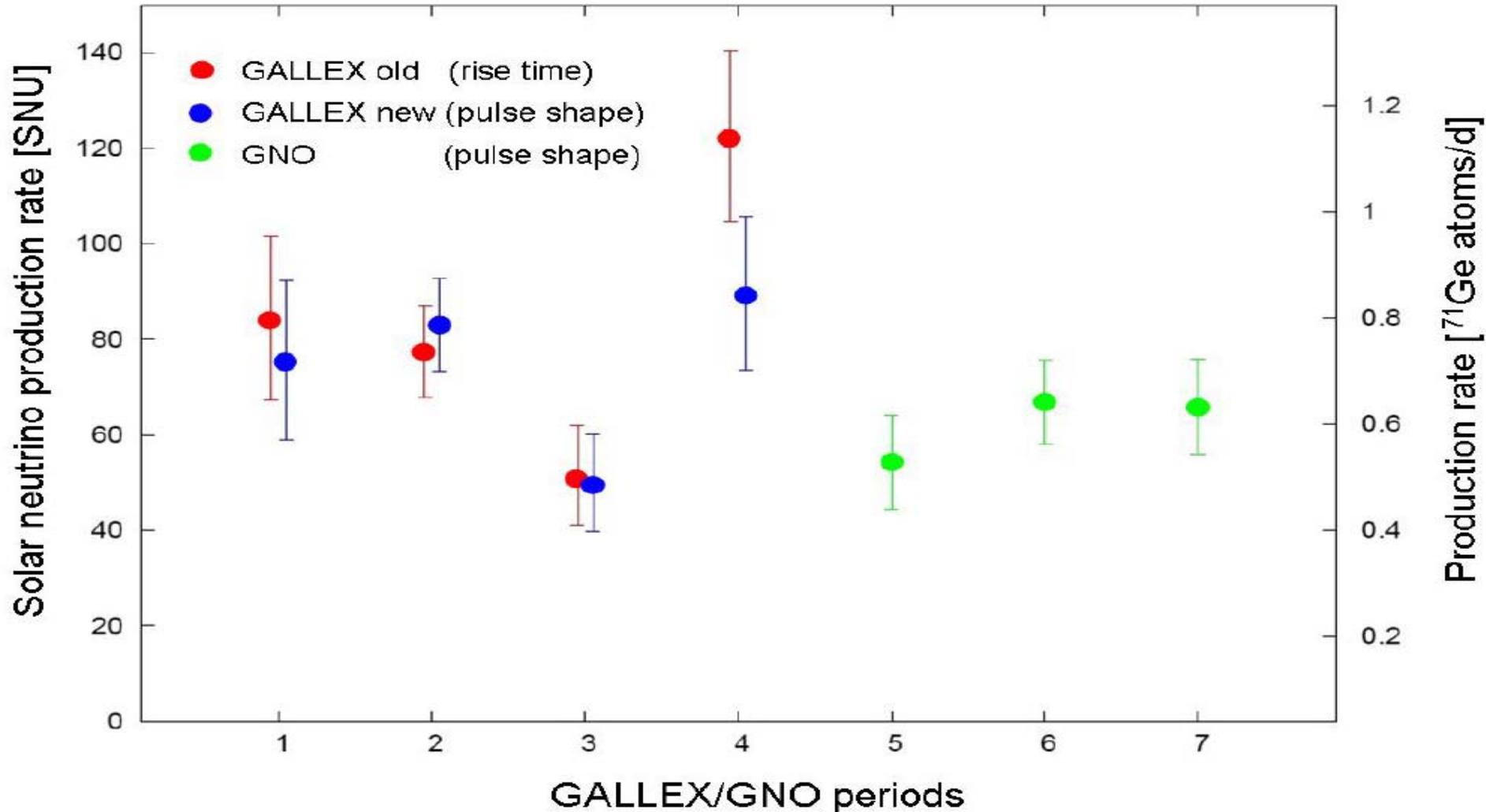


Updated SNU's from GALLEX (May 08)



Updated GALLEX and GNO Values

(May 08)



Updated Mean SNU Values from GALLEX (PSA) and GNO (May 08)

- GALLEX I: $75.1 + 17.3 - 16.2$
- GALLEX II: $82.8 + 10.0 - 9.5$
- GALLEX III: $49.5 + 10.7 - 9.8$
- GALLEX IV: $89.2 + 16.6 - 15.5$
- **GALLEX Combined Result (PSA) for I-IV:**
 $73.4 (+6.1 - 6.0) (+3.7 - 4.1)$
- GNO (I+II+III)₂₀₀₅: $62.9 (+5.5 - 5.3) \pm 2.5$
- **New GALLEX + GNO: $67.6 \pm 4.0 \pm 3.2$ (stat ~ syst)**
- **Range of SSM predicted rates:**

No oscillations **$122 - 131$ SNU**

With oscillations **$68 - 72$ SNU (global fit)**

Arsenic Tests in GALLEX

Made ^{71}Ge from $^{71}\text{As}^*$ Decay, not from ν 's

Cyclotron produced ^{71}As , added it to the Ga

Did multiple tests where varied the standing time, the chemical mixing and extraction conditions, method of Ge carrier addition

Purpose: to quantify any losses from chemical or 'hot-atom' effects

Did "triple-batch" comparison

$\approx 30\,000$ ^{71}As atoms added to:

Tank sample

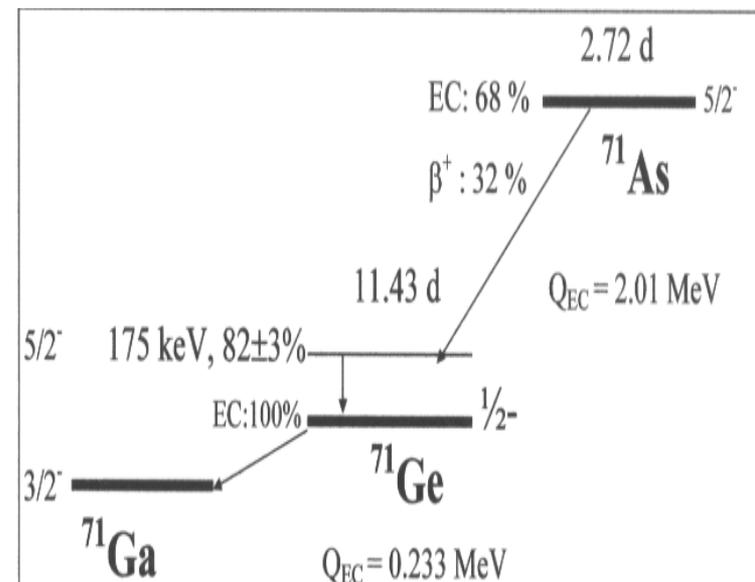
External sample

Calibration sample (γ -spectrum.)

Result: No Losses,

^{71}Ge Recovery 99+ %

The chemistry works!



GALLEX Cr- source update (PSA) (May 08)

- 2 source runs, S1, S2. Compared the measured ^{71}Ge to the ^{71}Ge expected from the known ^{51}Cr decay rate.

Mean S1+S2: **93** \pm 8% (1σ) Gallex PL (1998)

Re-evaluated Mean: **88.2** \pm 7.8% (1σ) Thesis Kaether (2007)

from S1: 95.3 \pm 11%, S2: 81.2 \pm 11%

- Value is $< 1\sigma$ from the expected 95 \pm 1% contribution from the ground-state-only transition Bahcall PR C (1997)
- So, the excited state contribution is probably close to 0, instead of (5 \pm 3)% as estimated by Bahcall
- Conclusion supported also by the SAGE Cr and Ar source results
- Note: GALLEX ^{71}As -experiment excludes Ge-yield errors $> 1\%$

The End of Gallium at Gran Sasso

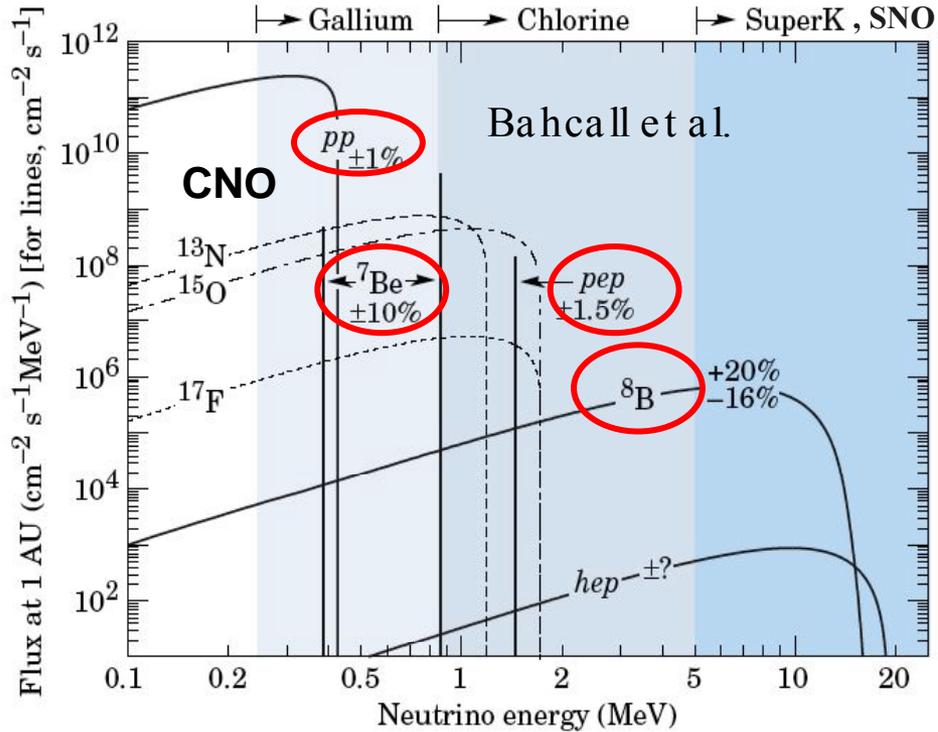
Febr 28, 2006: *Final Celebration Ceremony* for GALLEX/GNO at Gran Sasso, ending a successful fifteen year period that started with the Inauguration Ceremony on November 30, 1990

GALLEX was decommissioned and dismantled.

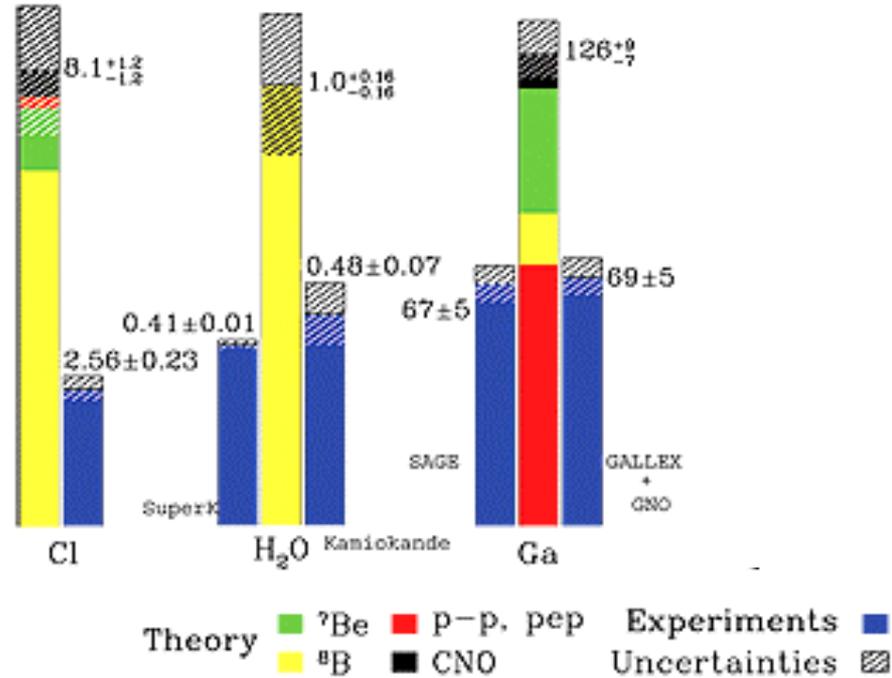
The gallium was removed from LNGS; later was sold, in April 2007

Flavour Change for Solar Neutrinos

Solar Model Flux Calculations



Previous Experiments Sensitive to Electron Neutrinos



SNO was designed to observe separately ν_e and all neutrino types to determine if low ν_e fluxes come from flavor change or solar models

Conclusions – I

- The radiochemical Cl and Ga experiments have been important contributors to the advances in our understanding of ν properties, and in solving the SNP
- In the early 1990's, they and Kamiokande were the only operating neutrino experiments
- However, the **radiochemical experiments** operate in batch mode,
 - a) yielding only one physical quantity, the SNU (or production) rate, which is proportional to the solar ν flux.
 - b) and which must be interpreted in terms of the SSM
- Contrast with the **real-time detectors** that see the neutrino interactions event by event,
 - a) yielding several neutrino parameters, such as the ν spatial distribution in the detector, the energy spectra, directionality, and even the oscillation pattern
 - b) SNO, by detecting both the CC and NC interactions, provided proof of flavor oscillations independent of the SSM,

Conclusions – II

- Real-time ν detectors have made consistent progress in lowering their detection thresholds, from ~ 9 MeV for Kamiokande to ~ 1 MeV for KamLAND and ~ 0.3 MeV for Borexino. Also see LENS, CLEAN, e-Bubble... for the solar pp region,
- As we have seen, SAGE will continue to run, perhaps even until the new real-time pp detectors will become realities
- **In my career, I have seen a natural progression in which nuclear chemical methods, where one observes the radioactive products of nuclear interactions after the interactions have occurred, have been supplanted by real-time detection of diverse nuclear phenomena, not only of neutrino reactions but also for example of nuclear fission and of complex heavy-ion high-energy nuclear reactions, such as those at RHIC.**
- In view of this, **I do not see that much incentive exists for developing new radiochemical ν detectors; certainly none appear imminent.**

Conclusions – III

- However, I do think that (nuclear) chemists will continue to play a significant role in ν research, e.g.,
 - a) in developing new detector systems, such as metal-loaded liquid scintillators, cryogenic detectors, ...
 - b) in detecting and reducing the levels of radioactive contaminants, such as U, Th, Ra, Rn, K...
 - c) developing new radioactive neutrino calibration sources
 - d) studying the long-term chemical interactions and compatibility of new detector substances with detector construction materials, such as the detector containment vessels
 - e) being concerned about chemical safety issues...
- The bottom line is that development of new ν detectors requires expertise in several scientific disciplines

>40 Years of Neutrino R&D @ BNL Chemistry Dep't.

Nuclear and Radiochemistry

- **Done:** **HOMESTAKE** Radiochemical Detector



- **Done:** **GALLEX** Radiochemical Detector



- **Done :** **SNO** Water Čerenkov Real-time Detector

Ultra-pure D_2O (1996 - \geq 2006)

- **New :** **#1 Focus - THETA-13**

High-Precision Experiments at [Daya Bay Nuclear Reactors](#)

Real-time Detector (R&D) Gd in [Liquid Scintillator](#), [Gd-LS](#) (began 2004)

- **New:** **SNO+** Real-time Detector (R&D) at **SNOLab**

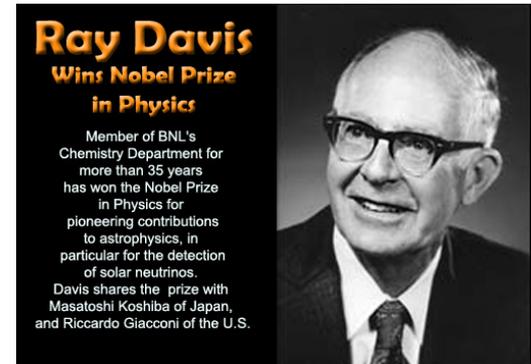
- ${}^{150}\text{Nd}$ -[LS](#) (began 2005) Double beta-decay

- **New:** **LENS, MiniLENS** Real-time Detector (R&D)

- ${}^{115}\text{In}$ -[LS](#) (began 2000), Detect pp and ${}^7\text{Be}$ Solar Neutrinos

- **New:** **Very Long-Baseline Neutrino Oscillations**

- ν_μ Beam from Accelerator to DUSEL (R&D began 2002)



Note: Hahn became Leader of BNL Group in February 1987, the same time as SN-1987A