A PROGRAM OF RADIOCHEMICAL SOLAR NEUTRINO EXPERIMENTS

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I. Introduction

The measured ^{37}Ar production rate in the Homestake chlorine solar neutrino detector, based on approximately five measurements each year since 1970, is 2.1 $\pm$ 0.3 SNU (1 solar neutrino unit = 10^{-36} captures $sec^{-1}atom^{-1}$) above a terrestrial background of 0.4 SNU. The most recent calculations based on standard solar models and the latest nuclear physics data suggest an expected rate of 6 - 7 SNU, a factor of roughly 3 above the measured results. The chlorine experiment is sensitive primarily (74% of the signal) to the high-energy ^{8}B solar neutrinos which comprise only about 10^{-4} of the total expected flux from the sun, and whose flux depends critically on the details of the interior structure of the sun (in particular, the central temperature).

By contrast, the bulk of the signal in a gallium detector (57%) arises from the primary p-p neutrinos. Since the proton-proton reaction is the essential first step in the hydrogen fusion chain, the flux of these neutrinos (6.1×10^{10} $cm^{-2}sec^{-1}$) is a constant calculated directly from the solar luminosity, provided only that the solar output is constant in time and that the sun's energy arises from hydrogen fusion. Finally, a bromine detector provides data on the intermediate-energy ^{7}Be neutrinos. Therefore the gallium experiment can be expected to provide a direct test of the presence of nuclear fusion processes in the sun's core, and the combination of gallium, bromine, and chlorine provide spectral data with the unmatched low background attainable only with radiochemical detectors.

In this paper, we describe a program of radiochemical solar neutrino experiments to be carried out as a collaboration between the Institute of Nuclear Research of the Academy of Sciences of the U.S.S.R. and the University of Pennsylvania. A 60 ton gallium detector and a 3000 ton ^{37}Cl detector will be constructed in the Baksan laboratory of the INR, and the present 615 ton Homestake chlorine detector will be converted into a 1000 ton bromine experiment. All three detectors will then be operated simultaneously.

II. The Homestake Chlorine Detector

The ^{37}Cl detector in the Homestake Mine[1] consists of 615 tons of tetrachloroethylene (C_2Cl_4) at a depth underground of 4850 ft (4000 meters water equivalent) and uses a radiochemical technique based on the reaction

$$\nu_e + {}^{37}Cl \longrightarrow {}^{37}Ar + e^- \quad .$$

In the following, we briefly describe the chemical methods, electronics, and background reduction and estimation procedures, not only because the chlorine results provide a major incentive for the gallium experiment, but also because the experience acquired in counting ^{37}Ar at very low background levels has been extremely valuable in establishing similar procedures for counting ^{71}Ge.

Procedure. The chemical procedures for removing argon are relatively straightforward. Helium gas is pumped through the liquid and then circulated in series through a condenser, a molecular sieve to trap residual C_2Cl_4 vapor and a liquid nitrogen-cooled charcoal trap to collect the argon gas. The argon collection efficiency is in excess of 95%. The argon is then purified and inserted into a small (4 mm inner diameter x 20 mm long) proportional counter which is placed in turn inside a large (30 cm diameter x 20 cm thick) sodium iodide anticoincidence shield. The sodium iodide scintillator is housed inside a low-background counting assembly consisting

of 30 cm of lead, 5 cm of boron-loaded polyethylene, and 20 cm of mercury. Care is taken to use low-radioactivity materials in the counters, in particular ultrapure quartz for the envelope. As a result, even without using the sodium iodide veto, the total 1 - 5 keV rate in the proportional counter has been reduced to less than 0.5 day^{-1}.

The electronics discriminate effectively against background by measuring both the pulse amplitude and rise time. The Auger electrons from the ^{37}Ar decay produce a 2.8 keV pulse. Since the Auger electron range is roughly 0.05 mm, the energy deposition is very localized, resulting in a characteristic fast-rising pulse. Background betas and Compton electrons typically have ranges equal to or greater than the counter diameter, and thus give rise to background counts with longer rise times and pulse heights spread out over the entire energy range. Over the last twelve runs the average background rate has been 0.007 day^{-1}.

Using a maximum likelihood technique[2], the observed events are separated statistically into a 35-day decay component and a constant background, taking into account the statistics of argon production, extraction, and counting, and arriving finally at a most likely value for the ^{37}Ar production rate in the detector.

Results. Figures 1 and 2 show the individual experimental values over the period 1970 - 1985, and also the yearly values. The average ^{37}Ar production rate over the entire period is 0.47 $\pm$ 0.04 ^{37}Ar atoms day^{-1} in 615 tons of C_2Cl_4. Allowing for a background ^{37}Ar production rate of 0.08 $\pm$ 0.03 atoms day^{-1} from cosmic rays gives an ^{37}Ar rate above estimated background of 2.1 $\pm$ 0.3 SNU.

Background Studies. Radiochemical detectors are relatively insensitive to extraneous electrons produced by internal contaminants, external gamma rays and neutrons from the rock walls, and cosmic rays. On the other hand, these same backgrounds typically limit direct counting techniques (water Cerenkov, scintillation, or track projection chambers) to neutrino interactions releasing electrons with 5 MeV or more. Only radiochemical detectors have been demonstrated to have the low thresholds and background rejection capabilities required to measure the low-energy p-p and ^{7}Be neutrino fluxes. Internal contaminations from uranium, thorium, and radium are easily controlled by analysis and screening of the materials in the chlorine (and the gallium and bromine) detectors, and the small effect of fast neutrons from the rock walls can be suppressed by surrounding the detector with a water shield. As described above, the non-^{37}Ar counter background in the ^{37}Cl detector has been reduced to a very low level by the use of shielding

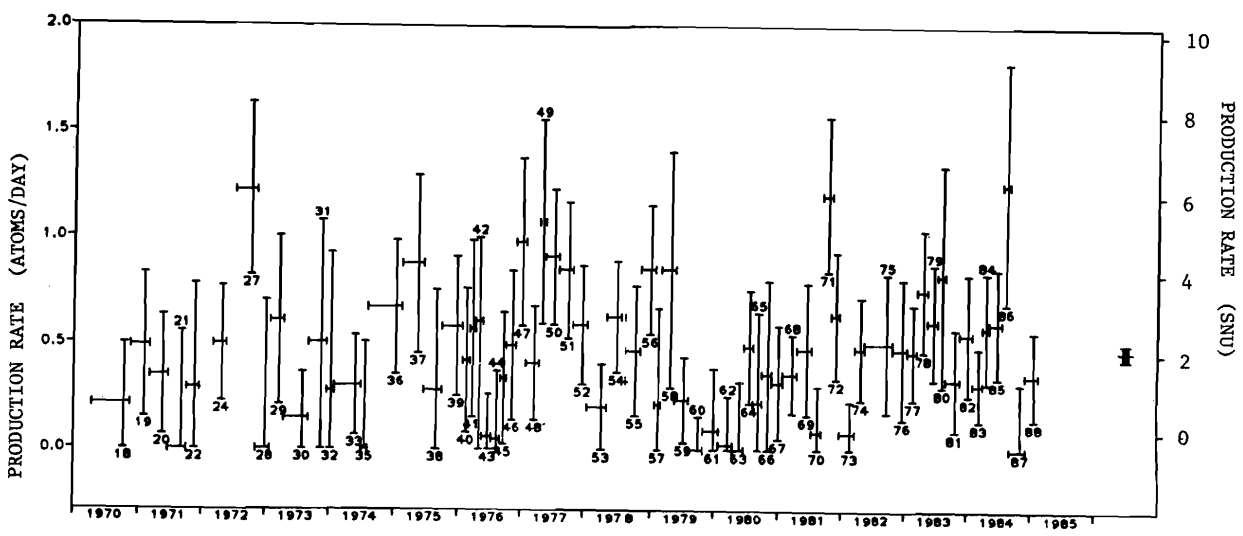

Fig. 1 Measured ^{37}Ar production rates for 67 individual runs from 1970 through the start of 1985, together with the 1σ error estimates.

and electronic measures.

The largest remaining source of background is due to the small remnant flux of atmospheric muons that give rise to secondary low-energy protons and pions which are then captured on ^{37}Cl to produce ^{37}Ar: $^{37}Cl(p,n)^{37}Ar$. A direct measure of the cosmic ray-induced background has been determined by measuring the ^{37}Ar production rate in small C_2Cl_4 detectors at shallow depths and extrapolating to 4000 m.w.e; these results agree well with analytic calculations[3] and with the results of muon beam measurements, and suggest a background rate of 0.075 ^{37}Ar day^{-1} in the full chlorine detector.

Fireman et al[4] have measured the muon-induced background using the reactions $^{39}K(X,Xpn)^{37}Ar$ and $^{39}K(X,Xnn)^{37}K(e^+,\nu)^{37}Ar$ in ^{39}K, where X is the incident muon or the energetic cascade particle. Since these are primarily direct muon reactions, the yield of ^{37}Ar is far higher than in the secondary (p,n) reaction on ^{37}Cl. By measuring the ^{37}Ar rate in potassium at the 4000 m.w.e. level and also at shallower levels, and comparing to the rates in chlorine detectors at the same shallower levels, the background rate can be determined independently of theoretical models for the extrapolation and independently of inaccuracies in the knowledge of the depth and detailed rock composition. The

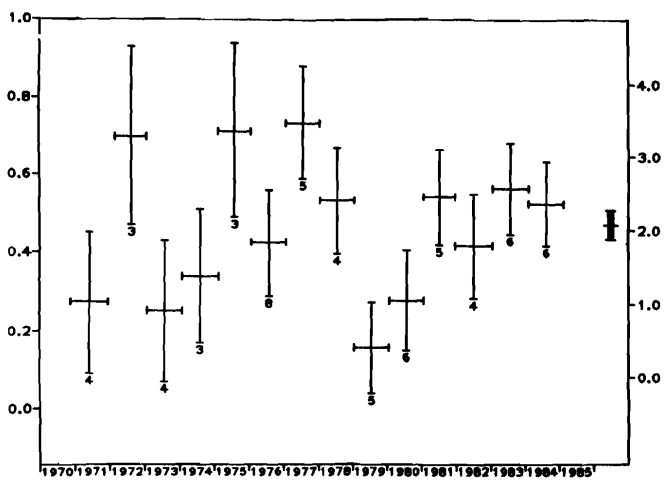

Fig. 2 Yearly average counting rates. The number below each point shows the number of individual runs represented by the yearly average.

initial results suggest a muon-induced ^{37}Ar production rate in the 615 ton chlorine detector of 0.07 ± 0.07 ^{37}Ar day^{-1}; the error in the potassium background figure will be reduced to about 20% within another year.

These background measurements are directly applicable to other radiochemical detectors. In particular, once the depth dependence of the ^{39}K detector is known, we can determine the muon-induced background in the gallium detector by carrying out a similar measurement with a small (1 ton) gallium detector at a suitable depth.

It is also important to note that background effects both from cosmic ray muons and internal contaminants can be measured directly in the ^{71}Ga and ^{81}Br detectors during the solar neutrino exposures. In the gallium experiment, ^{69}Ge (39 hr half-life) is more efficiently produced by cosmic ray muons than is ^{71}Ge (11 days), and (in gallium chloride) ^{37}Ar (35 days) is produced by alpha contamination. Similarly, in a bromine experiment, ^{79}Kr (35 hrs) is produced both by muons and internal contamination. Measurements of these background-produced activities can then be performed concurrently with the solar neutrino measurements, and provide a valuable check on the experiments.

III. The 60 Ton Baksan Gallium Experiment

As it became apparent that the results from the chlorine experiment were valid, and that the measured solar neutrino capture rate was too low to be explained by the standard solar model or any reasonable modifications, it also became clear that another solar neutrino experiment was needed -- one which would be as independent as possible of the details of the model calculations. Solar neutrino experiments are perhaps the only direct means of studying the sun's deep internal structure. In particular, the reaction $^{71}Ga(\nu,e)^{71}Ge$ suggested by Kuzmin[5] provides the basis for the only currently feasible experiment for observing the low-energy p-p neutrinos. Two gallium detectors are currently under construction: a 30 ton acid gallium chloride experiment in the Gran Sasso tunnel in Italy (GALLEX), and a 60 ton gallium metal detector

in the Baksan Valley in the USSR. The germanium extraction techniques for both the $GaCl_3$ and the gallium metal detectors were developed at Brookhaven between 1975 and 1978, and both provide satisfactory results[6]. The GALLEX detector has been described elsewhere[7]. Here we describe the Baksan experiment[8].

Germanium Extraction. Gallium metal is a dull grey material with a melting point of 29.5 C and a density of 6. Its low melting point makes it possible to keep it in its liquid form in the laboratory, and is central to the gallium-germanium separation. Stirring gallium in the presence of a 6.9% hydrogen peroxide solution with 1.7% hydrochloric acid (70 kg/7 tons) causes the gallium to break up into small globules whose surface layers are oxidized by the peroxide. The germanium preferentially migrates to the surface of the globules and is rapidly oxidized. A 22% HCl solution is then added to the system to dissolve the germanium and gallium oxides.

When the stirring is halted, the gallium metal and the hydrochloric acid separate gravitationally. The acid solution is then removed and reduced in volume by evaporating water. The evaporation continues until the acid concentration is approximately 5 molar. At that point HCl is added, increasing the acid molarity to 9 molar; $GeCl_4$ can then be swept out of the acid by a flow of argon (or other suitable gas) and subsequently trapped in a water scrubber. The remaining acid solution is separated into $GaCl_3$ and HCl. The HCl is reprocessed and recycled for the next extraction run. The $GaCl_3$ is stored and eventually converted back into gallium metal by electrolysis.

The final chemical steps are to further concentrate the $GeCl_4$ and convert it into a form suitable for counting (germane, GeH_4) by adding sodium borohydride. The extraction of $GeCl_4$ from the acid, and all subsequent procedures, are the same for the metal and gallium chloride detectors, and have been thoroughly developed and tested in prototype gallium metal and chloride studies[6]

In the 1975 experiments at Brookhaven, the effective extraction time from gallium metal was about 20 minutes with various prototype systems of 2 - 50 kg. At INR this procedure has been extended to a full-scale 7 ton detector module with the same extraction time[8]. Ten such modules, each containing 6 tons of gallium, will be used in the full 60 ton detector. In the U.S. prototype work, about 1% of the gallium was oxidized per extraction. Based on the experience with the 7 ton INR prototype, the gallium oxidation loss has been reduced to 0.1% per cycle with a germanium extraction efficiency of 91 $\pm$ 3%.

At a flux level of 124 SNU, 2.1 atoms o ^{71}Ge are produced in a 60 ton detector pei day, yielding 25 extracted atoms after a 3 week exposure (2 decay half-lives). Although this production rate is roughly 4 times higher than the ^{37}Ar rate in the ^{37}Cl detector, the counter background must still be extremely low.

One principle concern in the germanium extraction and concentration process is to avoid the introduction of contaminants into the extracted sample. In particular, HCl frequently contains germanium impurities. In the Baksan system, all materials will be recycled, thereby avoiding the introduction of fresh reagents and contaminants into the system. The first several extraction cycles will be dedicated to the removal of all extractable impurities from the system.

Tritium poses a particular problem. The tritium beta spectrum has an endpoint of 18 keV, so that the 10.4 keV Auger electron peak from ^{71}Ge decay will be contaminated by the presence of tritium betas. It is essential that tritium be completely removed from all reagents. This is accomplished by extracting the dilute $GeCl_4$ solution into tetrachloro-ethylene or carbon tetrachloride. The resultin solution is then dried to remove all water, yielding $GeCl_4$ in a hydrogen-free medium. From this point in the purification process, only tritium-free reagents, water (available from deep wells) and $NaBH_4$ will be used. Since commercial $NaBH_4$ contains tritium, a special tritium-free $NaBH_4$ must be prepared for use in the germane production. This is a tedious task

which is feasible but not yet successfully accomplished. One of the major chemical tasks in the experiment will be to produce the required supply of tritium-free $NaBH_4$ and to test the final stages of purification. This final low-level testing will be performed using the excellent existing rise time counting system in the Homestake Mine.

Non-radioactive germanium in the form of a Ga-Ge alloy is introduced into the gallium metal at the start of each run. The germanium is then recovered in the form of GeH_4 gas whose volume must then be accurately measured just before it is introduced into the counter. It is extremely important that the overall recovery be quantitatively accurate. In the gallium chloride pilot tests at Brookhaven, germanium recovery was measured with a precision of 1%. The gallium metal procedures used in the Baksan experiment present some challenges because of the two-phase metal-water solution and the complicated distillation and recovery procedures, but these procedures have been satisfactorily proven in the pilot studies.

Electronic Counting and Readout System. An extremely effective low-level risetime and pulse height electronic recording system was developed in connection with the chlorine experiment. Two systems, each capable of recording ^{37}Ar decay events from four individual counters, are currently in service at the Homestake Mine. The event rate is exceedingly low, the counters are stable over long periods of time (1 year), and the recording system is well-designed and reliable. The counters are calibrated with an x-ray source every two months. Otherwise, the system operates unattended in the mine.

A similar system for the Baksan installation will incorporate the following features:

1. The miniature gas proportional counters used for counting the ^{71}Ge electron capture decay are of a special design, and constructed of clean, selected materials. The counter envelope is fabricated from extremely pure fused silica (suprasil), and the cathode is made of selected pure iron or from an evaporated metal film. Special care must be taken in the counter design to minimize counter capacitance.
2. Improved designs for the timing amplifier, with lower noise levels and faster integration and differentiation periods, have recently been developed, and considerable development work has been devoted to pulse stretchers that can be directly applied to ^{71}Ge counting.
3. A computer-based logic system, incorporating a fast wave-form digitizer to record the ^{71}Ge pulse shape, will be used to record the data and perform the functions of the current hard-wired system.

Expected Results. The expected neutrino capture rates, based on the standard model calculation of Matthews et al.[9], are shown in Table 1. The total ^{71}Ga capture rate, assuming that the sun's core is currently producing energy by the p-p reaction at the rate needed to account for the surface luminosity, and assuming that neutrino oscillations can be neglected, is 124 SNU. Of this, 70 SNU come directly from the p-p reaction. Bahcall et al.[9] give an expected ^{37}Cl rate of 5.8 SNU, as compared to the measured ^{37}Cl rate of 2.1 SNU. If the suppression of the ^{37}Cl rate from 5.8 to 2.1 SNU is due to neutrino oscillations in vacuum, then we might expect a ^{71}Ga rate of 124 x (2.1 / 5.8) = 45 SNU. If the suppression is due to the Mikheyev-Smirnov-Bethe[10] version of matter oscillations, then only the high-energy ^{8}B neutrinos are suppressed (by a factor of 3), and the ^{71}Ga rate is 116 SNU, probably indistinguishable from the rate expected in the absence of oscillations. Other scenarios, assuming different choices for the mixing angle and neutrino eigenstate mass differences, result in correspondingly different rates for the gallium, bromine, and chlorine detectors, and are described by Beier et al[11] elsewhere in these proceedings.

With a flux of 124 SNU, the ^{71}Ga capture rate becomes 2 day^{-1}, as compared to the ^{37}Cl rate of 0.5 day^{-1}. With such a high rate, it will be possible to identify the sun as the source of the observed neutrinos by measuring

the annual changes in the counting rate as a function of the earth-sun distance[8]. Ten years of data with a 60 ton gallium detector should be capable of distinguishing the neutrino capture rates at perihelion and aphelion at the 87% confidence level and thus demonstrating the solar origin of these neutrinos.

Source Calibration Experiment. The response of the gallium detector to low energy neutrinos can be calibrated with an artificial ^{51}Cr source of monoenergetic 0.75 MeV neutrinos. In 1980, tests were carried out at Oak Ridge to determine the maximum possible source strength that could be produced at the HFIR reactor. Unfortunately, the neutron flux in the Cr target was insufficient to produce the MCi source required to test a $GaCl_3$ detector[12]. In the current experiment, this problem will be solved by using an isotopically-enriched sample of ^{50}Cr containing 87.7% ^{50}Cr compared to the normal 4.35% abundance of ^{50}Cr. The chromium will be dispersed throughout the reactor in order to avoid local reactor loading. This new approach should result in a sufficiently intense source to permit the calibration to be carried out.

Another factor favoring the source calibration experiment at Baksan is the fact that the density of gallium metal (ρ = 6 g/cm^3) is ten times that of gallium in the gallium chloride solution (ρ = 0.6 g/cm^3). Since the neutrino flux from the source falls off as r^{-2}, the ^{71}Ge production rate depends strongly on the gallium density near the source. A Ga metal target has a neutrino interaction rate almost an order of magnitude

Table 1: P-P Chain and C-N Cycle
Forecasted neutrino fluxes from the standard model and neutrino capture rates in ^{37}Cl, ^{71}Ga, and ^{81}Br.

Solar Reaction	Energy (MeV)	Flux ($10^{10}cm^{-2}sec^{-1}$)	Neutrino Capture Rate (SNU, from Standard Model[17]) ^{37}Cl	^{71}Ga	^{81}Br
Proton-Proton Chain					
$H+H \rightarrow D+e^{+}+\nu$	0-0.420	6.1	0	70.2	0
$H+e^{-}+H \rightarrow D+\nu$	1.44	0.015	0.024	3.0	1.2
$H+D \rightarrow {}^{3}He+\gamma$	---	---	---	---	---
${}^{3}He+{}^{3}He \rightarrow {}^{4}He+2H$	---	---	---	---	---
${}^{3}He+{}^{4}He \rightarrow {}^{7}Be+\gamma$	---	---	---	---	---
${}^{7}Be+e^{-} \rightarrow {}^{7}Li+\nu$	0.862,0.384	0.36	0.9	31.2	10.6
${}^{7}Be+H \rightarrow {}^{8}B+\gamma$	---	---	---	---	---
${}^{8}B \rightarrow e^{+}+\nu+{}^{8}Be^{*}$ $\rightarrow 2{}^{4}He$	0-15.0	0.00040	4.3	11.6	2.2
Carbon-Nitrogen Cycle					
$H+{}^{12}C \rightarrow {}^{13}N+\gamma$	---	---	---	---	---
${}^{13}N \rightarrow {}^{13}C+e^{+}+\nu$	0-1.2	0.05	0.08	3.3	1.0
$H+{}^{13}C \rightarrow {}^{14}N+\gamma$	---	---	---	---	---
$H+{}^{14}N \rightarrow {}^{15}O+\gamma$	---	---	---	---	---
${}^{15}O \rightarrow {}^{15}N+e^{+}+\nu$	0-1.73	0.04	0.24	4.6	1.6
$H+{}^{15}N \rightarrow {}^{12}C+{}^{4}He$	---	---	---	---	---
Total Neutrino Capture Rate			5.8	124	16.6

larger than a gallium chloride detector of the same mass[8].

We would like to make clear that a ^{51}Cr source primarily tests the ground state transitions. This state is well characterized and therefore can be accurately calculated (± 5 %). Values of cross-sections feeding the other excited states in ^{71}Ge (and in ^{81}Br) are determined by (p,n) reaction studies and nuclear model calculations[9,13]. These techniques are inherently not as reliable as the direct measurements of product nucleus lifetimes used to evaluate the cross-sections for the ground state transitions. By contrast, the ^{37}Cl cross-sections to all relevant excited states are well known from studies[13] of the mirror decay ^{37}Ca - ^{37}K.

IV. The 3000 Ton Baksan Chlorine Detector

The planned Baksan chlorine detector will contain 3000 tons of tetrachloroethylene, five times more than the present Homestake experiment. There are strong scientific reasons for building a larger chlorine experiment[1]. The data from the Homestake detector suggest an anti-correlation with the solar activity cycle as measured by sunspot occurrences and a correlation with coronal hole areas and the diameter of the sun. In addition, correlations have been seen between the ^{37}Ar rate and large solar flares[1]. There have been suggestions that these effects may be related to the existence of a neutrino magnetic moment[14]. Experimentally the effects are not yet conclusive and theoretically the discussions are still speculative, but the phenomenon clearly needs further study. Not only would the increased signal rate with a 3000 ton chlorine detector increase the significance of these apparent solar neutrino flux variations, but it would also permit the simultaneous observation of these variations at Homestake and at Baksan. It should be remembered that the chlorine detector is very sensitive to the internal dynamics and temperatures in the sun, and that the neutrino capture cross section is well known as a function of the neutrino energy.

Excavation of the chlorine chamber is scheduled to begin in early 1987, with the detector scheduled to be ready in 1990. Simultaneous observations will then be made with both the Homestake and Baksan detectors around the time of the next solar maximum.

V. The 1000 Ton Homestake Bromine Experiment

A bromine detector[15] based on the reaction $\nu_e + {}^{81}Br \longrightarrow {}^{81}Kr + e^-$ is primarily sensitive (Table 1) to the ^{7}Be neutrinos. The krypton is extracted from the detector using identical chemical techniques to those in the chlorine experiment. Preliminary extractions using the Homestake ^{37}Cl tank yield less than 400 ^{81}Kr atoms per sweep due to air leaks and other contamination. Because of the 2×10^5 yr half-life, the krypton atoms must be selectively ionized using resonance ionization spectroscopy and then mass selected with a quadrupole mass filter. If 500 ^{81}Kr atoms are extracted along with a background of 10^8 ^{82}Kr atoms, then two cycles of charge and mass selection are sufficient to yield 500 ^{81}Kr and 40 ^{82}Kr ions, which can then be counted as they are implanted in a Cu-Be target. At an expected level of 2 counts/day due to solar neutrinos and 0.1 background counts/day, extraction cycles must be 6 months - 1 year at the level of the present ^{81}Kr background limit.

The Homestake chlorine detector will be used for a full scale pilot test of the ^{81}Kr extraction and atom counting techniques. Extraction, purification, and detection of the ^{81}Kr by resonance ionization spectroscopy will be carried out using the Homestake detector with its present C_2Cl_4 filling and the addition of known quantities of ^{81}Kr. Since no bromine will be present, these tests can be used to determine quantitatively the ^{81}Kr recovery efficiency and detection sensitivity at the level of 100 - 1000 ^{81}Kr atoms, and will serve to demonstrate the ultimate sensitivity of the bromine detector under full scale conditions. When the Baksan chlorine detector has turned on and demonstrated that it can provide reliable answers, the chlorine will then be drained from the Homestake tank and replaced by a bromine compound. Even with the long extraction cycles,

the bromine will provide extremely valuable spectral information in conjunction with the Baksan chlorine and gallium results.

VI. The Baksan Laboratory and Schedule

The Baksan laboratory is located in the Caucasus Mountains near Mt. Elbrus. The mountain face rises abruptly from the floor of the valley, so that an overhead depth of 4000-4200 m.w.e. is obtained by driving a horizontal 3.5 km long adit into the mountain, where the gallium chamber complex has been constructed. The largest room, to be used for the gallium detector, is 60 m long x 12 m high x 11 m wide and lined with 60 cm of low radioactivity concrete and a 6 mm inner steel shell. Three additional rooms provide space for the germanium decay counting system and electronics; mechanical, electrical and air conditioning equipment; and offices. The gallium chamber construction is now complete and the room dehumidified. The installation of the ten gallium tanks has begun and should be completed by late fall 1986. The $GeCl_4$ extraction and conversion apparatus now in operation at Troitsk, the INR laboratory near Moscow, will be moved to Baksan early in 1987, as soon as the gallium installation is complete. Preliminary runs could begin as early as spring 1987. We anticipate that it will take much of 1987 to do system tests and eliminate any problems discovered during those tests. With luck, solar observations will commence by late 1987. Also in 1987 the adit will be extended 500 m and a room excavated for the 3000 ton chlorine experiment.

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17. For chlorine, we use the standard solar model calculations of Bahcall et al[9]; for gallium, we take the values from Matthews et al[9]; these are slightly higher than the corresponding values of Bahcall et al[9] because of the inclusion of excited state contributions. For bromine, we use the rates from Hurst et al[15].