Results from the Soviet-American gallium experiment

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RESULTS FROM THE SOVIET-AMERICAN GALLIUM EXPERIMENT


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ABSTRACT

A radiochemical $^{71}$Ga-$^{71}$Ge experiment to determine the primary flux of neutrinos from the Sun has begun operation at the Baksan Neutrino Observatory. The number of $^{71}$Ge atoms extracted from thirty tons of gallium was measured in five runs during the period of January to July 1990. Assuming that the extraction efficiency for $^{71}$Ge atoms produced by solar neutrinos is the same as from natural Ge carrier, we observed the capture rate to be $20 \pm 15/-20$ (stat) $\times 32$ (syst) SNU, resulting in a limit of less than 79 SNU (90% CL). This is to be compared with 132 SNU predicted by the Standard Solar Model.

1. Introduction

The discrepancy between the solar neutrino capture rate predicted by the Standard Solar Model (SSM) calculations and the $^{37}$Ar rate measured by the chlorine experiment in the Homestake Gold Mine has persisted for eighteen years. Recent calculated values of the flux are $7.9 \pm 2.6$ (3o) SNU (1 solar neutrino unit = $10^{-36}$ captures/target atom/s) in the Bahcall-Ulrich SSM and $5.8 \pm 1.3$ SNU (1$\sigma$) in the Turck-Chieze SSM. This is to be compared with the measured value in the chlorine experiment, averaged over the last eighteen years, which is $2.3 \pm 0.3$ SNU (1$\sigma$). This deficit has now been corroborated by the Kamiokande II water Cerenkov experiment, which observes only $0.46 \pm 0.05 \pm 0.06$ of the flux predicted by the Bahcall-Ulrich SSM, in fair agreement with the chlorine result.
The $^{37}$Cl and Kamiokande experiments are primarily sensitive to the high-energy $^8$B solar neutrinos, whose production rate depends critically ($T_{\odot}^{18}$) on the core temperature of the Sun. The neutrino spectrum, together with the thresholds for various detectors, is shown in Figure 1. Numerous nonstandard solar models\textsuperscript{5} that reduce the core temperature have been suggested, incorporating a variety of heavy element abundances, high magnetic fields, turbulent diffusion, continuous mixing, rapidly rotating or burned-out helium cores, convective mixing of hydrogen into the core, or new equations of state and other effects. However, none of the nonstandard models has been able to reproduce the observed $^8$B flux without running into difficulties accounting for other observed features on the Sun.

New particle physics, such as neutrino matter oscillations\textsuperscript{6-7}, neutrino magnetic moments\textsuperscript{8}, transition moments\textsuperscript{9}, neutrino decays\textsuperscript{10}, weakly interacting massive particles\textsuperscript{11} has been invoked, and nuclei with extra quarks\textsuperscript{12} in the solar core has been invoked to provide an explanation of the "solar neutrino problem".

A recent analysis\textsuperscript{13} of the consistency of the chlorine and Kamiokande II results concludes that the results are highly inconsistent with any astrophysical explanations and in fact are best attributed to Mikheyev-Smirnov-Wolfenstein (MSW) neutrino oscillations. The range of mass difference squared and mixing angles allowed by the chlorine and Kamiokande II results are shown in Figure 2.

The role new neutrino properties may play in the suppression of the high-energy solar neutrino flux, as possibly indicated in the chlorine and Kamiokande II experiments, can be determined from a measurement of the low-energy neutrinos produced in the dominant proton-proton (p-p) reaction. Exotic hypotheses aside, the rate of the p-p reaction is directly related to the solar luminosity and is insensitive to alterations in the solar models. An experiment using $^{71}$Ga (where the inverse beta decay reaction $\nu_e(^{71}\text{Ga},^{71}\text{Ge})e^-$ has a Q-
value of only 233 keV, allowing efficient detection of p-p neutrinos, which have an endpoint energy of 420 keV) as the capture material provides the only feasible means at present to measure low-energy solar neutrinos. The SSM calculations of Bahcall and Ulrich show that the dominant contribution to the total expected capture rate in $^{71}$Ga [132 + 20/-17 SNU (3σ)] arises from the p-p neutrinos [71 ± 4 SNU (3σ)]. Contributions by $^7$Be neutrinos (34 SNU) and $^8$B neutrinos (14 SNU) are also important. The insensitivity to variation in the SSM is seen in the calculated results from Turck-Chieze of 125 ± 5 SNU (1σ) for gallium. An observation in a gallium experiment of a strong suppression of the low-energy solar neutrino flux requires the invocation of new neutrino properties. The range of parameter space for neutrino oscillations allowed for different rates measured in a gallium experiment is indicated in Figure 2 by the dotted lines.

2. The Baksan Gallium Experiment

2.1. The Baksan Neutrino Observatory

In this paper we present the results of the first measurement of the solar neutrino flux by the Soviet-American Gallium solar neutrino Experiment (SAGE). SAGE uses the Gallium-Germanium Neutrino Telescope situated in an underground laboratory specially built at the Baksan Neutrino Observatory of the Institute for Nuclear Research of the Academy of Sciences of the USSR in the Northern Caucasus of the USSR. The main chamber of the laboratory is 60 m long, 10 m wide, and 12 m high. It is located 3.5 km from the entrance of a horizontal adit driven into the side of Mount Andyrchi, and has an overhead shielding of 4700 mwe. The laboratory is lined with a 6-mm steel shell and 60 cm of low-radioactivity concrete, in order to reduce neutron and gamma backgrounds from the rock.

2.2. Extraction Procedure

The extraction is based on the fact that gallium melts at 29.0°C. The low melting point makes it possible to keep the gallium in its liquid form and is central to the extraction. The gallium is contained in chemical reactors, each with internal volume 2 m$^3$ and lined with teflon. The reactors are provided with heaters that maintain the temperature just above the gallium melting point, and with stirrers to allow complete mixing of the extraction solutions with the gallium. Each reactor holds about 7 tons of gallium.
The chemical extraction process from metallic gallium was first worked out in the US\textsuperscript{15} and later fully tested in a 7.5-ton pilot experiment in the USSR\textsuperscript{16}. Each measurement of the solar neutrino flux begins by adding approximately 160 micrograms of natural Ge carrier in the form of a solid Ga-Ge alloy to each of the four reactors holding the gallium. The reactor contents are stirred so as to thoroughly disperse the carrier throughout the Ga metal. After a typical exposure interval of 3 to 4 weeks, the Ge carrier and any $^{71}\text{Ge}$ atoms that have been produced by neutrino capture are chemically extracted from the gallium using the following procedure.

Mixing gallium metal with a weak acidic solution in the presence of an oxidizing agent results in the extraction of germanium into the aqueous phase. The extraction process begins by adding to each reactor an extraction solution containing 1 kg of HCl, 5.2 kg of H$_2$O$_2$, and 68.8 kg of H$_2$O. The mixture is intensively stirred and the Ga metal turns into a fine emulsion. The total volume of the extraction solution from each reactor is 70 l. To ensure that the starting reagents are free of germanium to an acceptable level, the HCl solution (concentration 7N) and the H$_2$O are purified before use. Measurements of the H$_2$O$_2$ indicated sufficiently stringent limits on the presence of germanium that further purification was not deemed necessary. So as to minimize heating of the Ga during this strongly exothermic reaction, the HCl solution is cooled to $-15$ C and the water to 4 C. The Ge dissolved in the Ga migrates to the surface of the emulsion droplets. During this process, the Ge atoms in the gallium form GeCl$_4$. In approximately 10 minutes, the H$_2$O$_2$ is consumed; almost all of the emulsion spontaneously breaks down and the phases separate. The extraction procedure is then finished by adding 43 l of 7N HCl and stirring for 1 min. Less than 0.1\% of the gallium has been oxidized and the gallium temperature has risen to about 50 C. The extraction solution is then siphoned away from each reactor and the reactors are washed by adding 20 l of 0.5N HCl. This solution is vigorously stirred with the liquid gallium for about 1 min and is then siphoned away to be added to the previous extraction solution.

All of the extracts from the separate reactors are combined, and the Ge is then concentrated by vacuum evaporation in a glass apparatus. Since Ge is volatile from concentrated HCl solutions, the distillation is stopped when the volume has been reduced by a factor of four. The solution is then transferred to another glass vessel that is part of a sealed gas flow system. Purified 12N HCl is added to this solution to raise the HCl concentration to 9N, and an argon purge is initiated. The argon flow (1.0 m$^3$/hr for 1.5 hours) sweeps the Ge as GeCl$_4$ from this acid solution into a volume of 1.0 l of H$_2$O. When this process is completed, a solvent extraction procedure is used to first extract the Ge into CCl$_4$ and then back-extract it into H$_2$O. This process is repeated three times and the residual CCl$_4$ is removed by heating the water to 90 C for 1.5 hours. To improve the efficiency of CCl$_4$ removal, a very small amount of hexane is added to the organic phase at the last step of the final back-extraction.
The next step of the procedure is to synthesize the counting gas GeH₄. The synthesis reaction is optimized at a pH of 8-9, so NaOH is added to adjust the pH to this range. The resulting solution, with volume now only 100 ml, is placed in a small reactor flask in a sealed helium flow system. Here, 50 ml of a 0.02N NaOH solution containing 2 g of NaBH₄ is added. GeH₄ (germane) is produced when this mixture is heated to about 70°C. The helium flow sweeps the germane into a gas chromatography system where it is purified. A measured quantity of xenon is added, and this mixture is inserted into a sealed proportional counter with volume of 0.75 cm³ and counted for 2-3 months. To prevent gallium oxidation, 100 l of 1N HCl are added to each reactor when the extraction process is complete. This acid solution is discarded immediately before the next extraction. A measured quantity of xenon is added, and this mixture is inserted into a sealed proportional counter.

The standard procedure is to conduct three extractions in series within a period of 5 days without adding additional carrier to the reactors. The GeH₄ samples from each of these three extractions are then usually counted separately. In addition, "blank" runs are carried out before and after each of these three extractions by carrying out the final stages of the extraction process beginning with the extraction into CCl₄. This is to ensure that no additional activity is being removed from the extraction system. The efficiency of extraction of the germanium carrier is measured at several stages of the extraction procedure by atomic absorption spectroscopic analysis. The first samples are taken from the extraction solutions from each reactor. Other samples are removed after the germanium has been swept into 1.0 l of H₂O and after the back-extraction from CCl₄ into H₂O. The final determination of the quantity of germanium is made by measuring the volume of synthesized GeH₄. The major uncertainty in these measurements is in the amount of Ge carrier added to the reactors. The Ge concentration in the carrier slugs is determined by exhaustive extraction of Ge from several representative slugs using the same procedure as for Ge extraction from the large reactors. The error on the quantity of Ge carrier is estimated to be ±5%. The overall extraction efficiency is typically 80% with an uncertainty of ±6%.

2.3. Counting

The SSM predicts a production rate of 1.2 ⁷¹Ge atoms/day in 30 tons of Ga. At the end of a 4-week exposure period, an average of 16 ⁷¹Ge atoms will be present. Under normal conditions, there is a one-day delay between the end of exposure and the beginning of counting. Taking this delay into account and folding in the chemical extraction and detector counting efficiencies, only about 4 ⁷¹Ge atoms are expected to be detected in each run. Thus, the counting backgrounds must be kept to a small fraction of a count/day.

⁷¹Ge decays with an 11.4-day half life, by electron capture to the ground state of ⁷¹Ga. The probabilities of K, L, and M capture are 88%, 10.3%, and 1.7%, respectively. The only way to observe this decay is to detect the low-
energy Auger electrons and x-rays produced during electron shell relaxation in the resulting $^{71}$Ga atom. K capture gives Auger electrons with an energy of 10.4 keV (41.5% of all decays), 9.2-keV x-rays accompanied by 1.2-keV Auger electrons from the subsequent M-L transition (41.2% of all decays), and 13.26-keV x-rays accompanied by 0.12 keV Auger electrons (5.3% of all decays). L and M capture give only Auger electrons with energies of 1.2 keV and 0.12 keV, respectively.

These low-energy electrons are detected in a small-volume proportional counter. If xenon is mixed in with the germane to increase the probability of capturing some of the 9-keV x-rays, then a typical counter filled with an 80% Xe - 20% GeH$_4$ mixture at 600 Torr gives a resolution of 18-21% at 5.9 keV, and 37% of the decays are observed in the Ge K peak at 10.4 keV and 34% in the L peak at 1.2 keV. Due to considerably higher backgrounds in the L peak, only the K peak has been used in the analysis presented here.

The proportional counter (with a volume of about 0.75 cm$^3$) is placed in the well of a NaI detector inside a large passive shield and counted for 2-3 months. The NaI detector provides identification of any gammas associated with a pulse in the proportional counter. Typically, a 9" × 9" crystal is used, and the NaI count rate inside the passive shield above 200 keV is about 3 Hz.

Pulse shape discrimination based on rise-time measurements is used to separate the $^{71}$Ge decays from background. In contrast to the spatially localized ionization produced by Auger electrons or x-rays from $^{71}$Ge decay, background radioactivity primarily produces fast electrons in the counter, which result in extended ionization. Pulses from the counter are differentiated with a time constant of 10 ns. The amplitude of the differentiated pulse is proportional to the product of the amplitude and the inverse rise time of the pulse. For every event in the counter, the energy, the amplitude of the differentiated pulse, and any associated NaI signal are recorded.

2.4. Detector Calibration

After filling a counter with the GeH$_4$-Xe mixture from an extraction from the 30 tons of gallium, the counter is calibrated using an external $^{55}$Fe source, which illuminates the central part of the counter through a thin side window. Calibrations of the counters are repeated at approximately one-month intervals. The stability of the counters used in the data reported here was quite good, with typical gain variations observed of 3-4%.

The $^{55}$Fe calibration is used to generate a two-dimensional plot of inverse rise time versus energy. A rectangular acceptance window is then calculated around the 5.9-keV $^{55}$Fe peak, which accepts 95% in energy centered around the Fe energy peak, and 95% of the rise-time distribution. The position of the acceptance window for the $^{71}$Ge K peak is calculated by first determining any offsets using a linear pulser. The centroid of the acceptance boxes for the $^{71}$Ge K peak is then determined by scaling from the $^{55}$Fe peak.
The width of the $^{71}\text{Ge}$ K peak window is determined by scaling the width in energy as the square root of the energy and setting the width in rise time to be constant and equal to the width for the $^{55}\text{Fe}$ peak. In order to check this extrapolation procedure, a counter was filled with the standard GeH$_4$-Xe mixture in which the Ge had been activated in the Los Alamos Omega West reactor. This provided an internal calibration source of $^{71}\text{Ge}$ in the counter gas with a total counting rate of less than 10 counts/s. All of the counting systems were calibrated using this counter. The spectrum from this counter taken simultaneously with the external $^{55}\text{Fe}$ source is shown in Figure 3. The acceptance boxes for the $^{55}\text{Fe}$ and the $^{71}\text{Ge}$ K peak are marked and these data clearly show that the extrapolation method used is correct. The peaks occur all with the same rise time, since they are all due to low energy x-rays that produce point ionization in the counter. Events with lower values of $1/\text{rise time}$ are due to background pulses that produce extended ionization in the counter.

2.5. $^{71}\text{Ge}$ Background

The main source of $^{71}\text{Ge}$ in the reactors other than from solar neutrinos is from protons arising as secondary particles produced by i) external neutrons, ii) internal radioactivity, and iii) cosmic ray muons. These protons can initiate the reaction $^{71}\text{Ga}(p,n)^{71}\text{Ge}$. The use of metallic gallium (as opposed to Ga in an aqueous solution) results in reduced sensitivity to both internal and external backgrounds. Extensive work has gone into measurements and calculations of these background channels:

i) Since the (n,p) cross sections on the Ga isotopes are small and the laboratory has been lined with low-background concrete, the external neutron background in 30 tons of Ga metal has been calculated to produce no more than 0.01 atoms of $^{71}\text{Ge}$ per day.

ii) The background from internal radioactivity is mainly determined by the concentrations of U, Th, and Ra in the gallium. Measurements of these
ii) The background from internal radioactivity is mainly determined by the concentrations of U, Th, and Ra in the gallium. Measurements of these concentrations, combined with measured yields of $^{71}\text{Ge}$ from alpha particles\textsuperscript{15,18}, indicate that less than 0.01 atoms of $^{71}\text{Ge}$ will be produced per day in 30 tons of Ga metal.

iii) Based on the measured muon flux in the laboratory of $(2.4 \pm 0.3) \times 10^{-9}$ muon/cm$^2$/s, the production rates of the germanium isotopes from cosmic ray muons have been calculated\textsuperscript{19} to be 0.005 $^{71}\text{Ge}$, 0.013 $^{69}\text{Ge}$, and 0.009 $^{68}\text{Ge}$ atoms per day in 30 tons of Ga metal.

Another type of background that arises only during counting can come from tritium in the counting gas. In order to eliminate this source of counter background, special methods for synthesizing NaBH$_4$ have been developed\textsuperscript{20} using starting ingredients selected to have a low tritium content.

Thus, the total background production rate in the 30 tons of liquid gallium of all germanium activities from all sources has been calculated to be less than 2.5\% of the SSM production rate.

3. Extraction History

The experiment began operation in May of 1988, when purification of the 30 tons of gallium commenced. Large quantities of long-lived $^{68}\text{Ge}$ (half life = 271 days) produced by cosmic rays while the gallium was on the surface had to be removed. The decay of $^{68}\text{Ge}$ cannot be differentiated from those of $^{71}\text{Ge}$, as $^{68}\text{Ge}$ also decays by electron capture. The subsequent decay of $^{68}\text{Ga}$ from $^{68}\text{Ge}$ (half life = 1.14 hours) is by positron emission in 90\% of the cases. In a proportional counter with 5-mm cathode diameter filled with 90\% Xe and 10\% GeH$_4$, the $^{68}\text{Ga}$ decay gives an energy spectrum with a broad peak whose maximum is at about 1 keV. These $^{68}\text{Ga}$ decays can be identified to some extent by rise-time analysis of the counter pulse and by detection of a coincidence pulse in the surrounding NaI crystal. The $^{68}\text{Ge}$ activity in the first extraction from the 30 tons of Ga was 7700 counts/day in the Ge K peak.

The chemical extraction efficiency and $^{68}\text{Ge}$ counting rate were monitored during most of the initial extractions and showed that the $^{68}\text{Ge}$ rates tracked the chemical extraction efficiency for the first nine or ten extractions. However, beginning with the extractions in January 1989, it became clear that there was a source of long-lived residual activity that did not come down rapidly with further extractions. The level of this activity was about one count/day in the Ge K peak. Although the source of this activity has not been definitely identified, exhaustive testing showed that it clearly came from either the reactor vessels or the gallium, not from contaminants in the reagents. The most likely possibility is that there was some diffusion of long-lived $^{68}\text{Ge}$ from the original dirty gallium into the teflon li 1 ers. Further extractions continued to slowly reduce this background. The reactor has now undergone at least 20 extractions and in recent runs, the activity in the Ge K peak is less than 0.2 counts/day.
With the extractions beginning in April 1989, the analysis indicated the presence of several counts per run with a half-life reasonably consistent with $^{71}$Ge. However, as statistics from additional extractions in June, September, October, and December 1989 built up, the best fit to the half-life was determined to be $4 \pm 1$ days, which is consistent with radon rather than $^{71}$Ge. Subsequently, a large, ultra-low background hyperpure germanium solid state detector was installed at the experiment and used to assay all of the reagents. It was found that the water used in the final stages of the germane synthesis did contain some radon. In order to eliminate the radon, new extraction procedures were implemented beginning with the January 1990 extraction. These procedures included using old, tritium-free water and additional vacuum distillation of other reagents to remove radon. These procedures resulted in the elimination of the radon contamination in the extractions.

The data from the January run are shown in Figure 4.

![Figure 4](image-url)

Figure 4. a) Rise time versus energy for January extraction data, b) blowup of rise time$^{-1}$.

By the beginning of 1990, the backgrounds had been reduced to levels sufficiently low to begin measurements of the solar neutrino flux. Some residual radioactivity is still present that produces events in the energy range of 1-15 keV. As seen in Figure 4, these events are predominantly below 6 keV; they have both slow and fast rise-time components with some events in coincidence with a NaI detector.

4. **Measurement of The Solar Neutrino Flux**

4.1. **Statistical Analysis**

Results from measurements carried out in January, February, March, April, and July of 1990 are reported here. Earlier data taken during 1989 are not presented here due to the presence of radon and $^{68}$Ge residual
contaminations. The run during May of 1990 was unusable due to an electronic instability, and the run during June of 1990 was lost due to a vacuum accident.

The data analysis selects events that have no NaI activity in coincidence within the $^{71}$Ge K-peak acceptance window. A maximum likelihood analysis is then carried out on these events by fitting the time distribution to an 11.4-day half-life exponential decay plus a constant rate background. The total background rate of selected counters from 0.7-13.0 keV is approximately 2.0 counts/day, with typical rates in the $^{71}$Ge K peak of 0.10 counts/day.

The results of the maximum likelihood analysis are shown in Table 1.

<table>
<thead>
<tr>
<th>Extraction Date</th>
<th>Best Fit (SNU)</th>
<th>$Nw^2$</th>
<th>Probability (%)</th>
<th>68% CL (SNU)</th>
<th>90% CL (SNU)</th>
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</thead>
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<td>0</td>
<td>0.367</td>
<td>9</td>
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<td>118</td>
</tr>
<tr>
<td>Feb 28</td>
<td>39</td>
<td>0.310</td>
<td>13</td>
<td>83</td>
<td>142</td>
</tr>
<tr>
<td>Mar 29</td>
<td>90</td>
<td>0.035</td>
<td>96</td>
<td>175</td>
<td>276</td>
</tr>
<tr>
<td>Apr 20</td>
<td>0</td>
<td>0.060</td>
<td>81</td>
<td>94</td>
<td>174</td>
</tr>
<tr>
<td>Jul 24</td>
<td>0</td>
<td>0.250</td>
<td>19</td>
<td>149</td>
<td>275</td>
</tr>
<tr>
<td>Combined</td>
<td>20</td>
<td>0.223</td>
<td>23</td>
<td>35</td>
<td>60</td>
</tr>
</tbody>
</table>

The data from each of the five extractions are shown in Figure 5, which shows the integral plot of events versus time within the $^{71}$Ge K-peak acceptance window. In this figure, the value of the curve is incremented by one count every time an event occurs and thus shows the time distribution of $^{71}$Ge-like events. The best fit line to each data set is shown by the dashed line. The results of the maximum likelihood statistical analysis are shown in Table 1. The Smirnov-Cramer-Von Mises parameter $Nw^2$ provides a measure of the goodness of fit which is independent of the binning of the data. For this parameter, it is expected that 50% of the fits should have values greater than 0.119, and 50% less than 0.119. (In some sense, one can consider a $Nw^2$ value of 0.119 as being analogous to a $\chi^2$ value of 1.0.) The probability that a measurement would exceed the value of $Nw^2$ determined for each of the runs is also given in Table 1. Thus, for example, we find a statistical probability that 23% of the time we would obtain a worse fit to the combined data, assuming a background constant in time and a signal of 20 SNU (1 Solar Neutrino Unit = $10^{-36}$ captures/target atom/s).

4.2. Systematic Effects

The systematic uncertainties in the chemical extraction and counting efficiencies were typically 6% and 10%, respectively. These were added in quadrature together with uncertainties in the amount of gallium (0.5%), the exposure time (0.1%), the delay time between start of the extraction and the start of counting (0.5%), and dead time in the counting system (0.5%) to obtain
an overall uncertainty in the total efficiency of 14%, which corresponds to uncertainties of 5 SNU (68% CL) and 14 SNU (90% CL).

The uncertainty in background determination under the $^{71}$Ge decay curve due to possible time variations of the counter background was checked in a number of ways. Analyses were made by truncating the data set for events in the K-peak acceptance window event by event in time. In the case of fitting only to 33 days (3 half-lives) of counting, the best fit value increased only to 31 SNU and the limits increased to 45 SNU (68% CL) and 74 SNU (90% CL). We also fit the time distribution of the combined data set for events in the K-peak window with a first-order polynomial expansion in time of the background rate. We then generated Monte Carlo data sets using the upper 68% and 90% CL limits of the parameters from the fit. We analyzed the time-dependent Monte Carlo, data assuming that the background rate was constant in time and determined the maximal change in the best fit to limits set on the $^{71}$Ge rate to be 30 SNU (68% CL) and 35 SNU (90% CL). This corresponds to an uncertainty of approximately one event per run being assigned incorrectly to signal rather than background due to a possible time variation in the background.

Finally, we binned the data from each of the five extractions in 2-keV wide bins from 1 to 13 keV. Each of the resulting 30 sets of data was analyzed and $Nw^2$ values determined under the assumption that a) the background rate is constant in time, or b) the background rate increased in time at the rate determined by the polynomial expansion fit described above. The distribution of $Nw^2$ values from the data sets under these two assumptions was fit and a single $Nw^2$ value for each assumption was calculated. As shown in Figure 6, the fit to a constant background rate is very good, giving a $Nw^2$ value of 0.053, corresponding to obtaining a better fit only 14% of the time. The fit, assuming an increasing background rate, was very bad with a $Nw^2$ value of 1.129, corresponding to less than a 0.2% probability. However, we note that while this
analysis is consistent with a constant background, it assumes that the background in the K-peak acceptance window comes from the same sources as the background over the entire energy range. Thus, in order to use the full counting time and to minimize any assumptions, we have conservatively chosen to choose the uncertainties determined by the second method, and therefore assign an uncertainty to any possible time variation of the background to be 30 SNU (68%) and 35 SNU (90% CL).

Figure 6. $N w^2$ distributions for all runs binned in 2-keV energy bins assuming a) constant rate background and b) increasing background rate.

The uncertainty in extrapolating the energy and inverse rise-time cuts was estimated using a cut that includes all events not in coincidence with the NaI counter and with an energy above the lower edge of the K peak. The fit to the combined data with this cut gave a best fit of 0 SNU and limits of 45 SNU (68% CL) and 83 SNU (90% CL). The difference between these limits and those determined using the normal energy and inverse rise-time cuts was taken
as the systematic uncertainty in extrapolating from the $^{55}\text{Fe}$ calibrations. This results in uncertainties of 10 SNU (68% CL) and 23 SNU (90% CL).

5. Results

The results of the analysis are, assuming that the extraction efficiency for $^{71}\text{Ge}$ atoms produced by solar neutrinos is the same as that measured using natural Ge carrier:

$$^{71}\text{Ga Capture Rate} = 20 \pm 15/20 \text{ (stat) } \pm 32 \text{ (syst) SNU}.$$ 

Upper limits were determined by adding the statistical and systematic errors in quadrature and then adding this linearly to the best fit value. The upper limits are:

$$^{71}\text{Ga Capture Rate} < 55 \text{ SNU (68\% CL), } < 79 \text{ SNU (90\% CL).}$$

In terms of the total number of $^{71}\text{Ge}$ atoms observed, these values correspond to a best fit of 2.6 atoms observed in the five runs. The SSM predicts a production rate of 1.2 $^{71}\text{Ge}$ atoms/day in 30 tons of Ga, corresponding to a total 17.0 atoms expected for the runs reported here. In fact, if one assigns all events within the K-peak acceptance window during the first two $^{71}\text{Ge}$ half-lives to be signal with no background, one observes only 9 events total, compared to 13 events predicted by the SSM in this time period.

While all available information leads one to expect that the extraction efficiency for $^{71}\text{Ge}$ atoms produced by solar neutrinos should be the same as for the carrier, it is important to test this assumption. A test to search for possible losses in the extraction of $^{71}\text{Ge}$ atoms compared with the natural Ge isotopes was carried out in which the Ge carrier was doped with a known number of $^{71}\text{Ge}$ atoms. The doped carrier was added to one of the reactors holding 7 tons of gallium, three successive extractions were carried out, and the number of $^{71}\text{Ge}$ atoms in each extraction was determined by counting. Table 2 shows the results of this measurement, and indicates that the extraction efficiency of the natural Ge carrier and $^{71}\text{Ge}$ track very closely. The third extraction had a sensitivity of only 200 atoms detected due to electronic problems with one.

<table>
<thead>
<tr>
<th>Run</th>
<th>Carrier(mg)</th>
<th>$^{71}\text{Ge}$ atoms</th>
<th>Efficiency (%)</th>
<th>Carrier</th>
<th>$^{71}\text{Ge}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Amount Added</td>
<td>525 ± 26</td>
<td>6555 ± 359</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>1</td>
<td>410 ± 10</td>
<td>5188 ± 195</td>
<td>78 ± 4</td>
<td>79 ± 5</td>
<td></td>
</tr>
<tr>
<td>2</td>
<td>97 ± 2</td>
<td>1131 ± 107</td>
<td>84 ± 20</td>
<td>84 ± 26</td>
<td></td>
</tr>
<tr>
<td>3</td>
<td>21 ± 1</td>
<td>&lt;200</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Sum</td>
<td>528 ± 10</td>
<td>6519 + 222/-422</td>
<td>101 ± 5</td>
<td>99 ± 6/-8</td>
<td></td>
</tr>
</tbody>
</table>
channel of the counting system. The half-life of $^{71}$Ge in this extraction test was measured to be $11.0 \pm 2.4$ days, in good agreement with the known half-life of 11.4 days.

A more extensive series of tests was performed at our surface laboratory near Moscow during the development of the radiochemical procedure using a module similar to the present detector, which contained 7 tons of gallium. In these tests, various germanium activities produced by cosmic rays were extracted. These tests showed that the radiochemical procedures used are valid and indicated that the natural germanium carrier yield should effectively measure the extraction efficiency of neutrino-produced $^{71}$Ge. Several additional tests are planned, including an experiment using a neutrino source.

A suitable neutrino calibration source can be made using $^{51}$Cr, which decays with a 27.7-day half-life by electron capture, emitting monoenergetic neutrinos of 751 keV (90.2% BR) and 426 keV (9.8% BR). An intense $^{51}$Cr source can be produced by irradiating $^{50}$Cr in a nuclear reactor. A full-scale calibration run is scheduled for 1993 using a 1-MCi $^{51}$Cr source. An engineering test run with a lower-intensity $^{51}$Cr source was carried out during the fall of 1990. A primary difficulty for the test run was an extended scheduled shutdown of the production reactor in October 1990 for reactor upgrades. Thus, the test run was mounted in a period of only three months, which did not allow time to prepare everything in the same final form as planned for the full-scale calibration run. Nonetheless, it was decided that valuable operating experience would be gained by carrying out an engineering test run with a lower-intensity source.

A source was produced at the SM-2 reactor in Russia by irradiating 212 gm of 87.7% enriched $^{50}$Cr in a flux of $2 \times 10^{15}$n/cm$^2$/s. Due to time constraints, only part of the chromium was zone refined, while the remainder was chemically purified. The irradiated chromium was inserted into a stainless steel cylinder, which was to make a compact source of 74 mm diameter by 134 mm high. Thermal calorimetric measurements determined the $^{51}$Cr activity to have been approximately 350 kCi. The level of high-energy gamma long-lived radioimpurities was determined by measurements made with a high-purity germanium detector to be about 0.4%.

Five consecutive irradiations of approximately 12 tons of gallium were made. For the first three exposures, newly purified gallium was used, while the last two exposures used old gallium from the solar neutrino runs. The normal procedure used was to mix natural Ge carrier into two gallium reactors and then to pump gallium from these two reactors into a single reactor in which the stirrer had been replaced by a reentrant port for the chromium source. The gallium was typically irradiated for a period of 13 days. It was then pumped back to the two original reactors and normal extractions were then carried out on these two reactors. Any $^{71}$Ge produced was counted using the same counters and electronics as used in the solar neutrino runs. The chemical extraction
efficiency using this process was measured to be the same as that typical for the solar neutrino runs.

Due to the short preparation time for the test run, the new gallium could not be entirely purified of residual $^{68}$Ge and reagents were not able to be purified to the same purity level as used during previous solar neutrino runs. This resulted in backgrounds in the test run almost an order of magnitude higher than in the previous solar neutrino runs. The dominant components of the background were $^{68}$Ge and radon. While the runs indicated some apparent production of $^{71}$Ge, due to the high background and the presence of radon, it was not possible to determine a reliable quantitative measure of the production rate of $^{71}$Ge. Nonetheless, a great deal of valuable experience from handling a few-hundred-kiloCurie source and in determining the technical problems involved in a calibration measurement was obtained. With this experience in hand, we plan to carry out a full-scale calibration experiment with a 1-MCi $^{51}$Cr source and 20-25 tons of gallium in 1993.

6. Current Status and Future Plans

Useful solar neutrino data were not obtained after the July run due to work on the chromium engineering test run. Following the completion of the test run, a total of about 30 tons of new gallium has been purified to remove $^{68}$Ge (including further purification of the gallium used in the chromium engineering test run). At the same time, the old gallium used in the previous solar neutrino runs was removed and the chemical reactors were extensively cleaned. The entire chemical extraction system was also carefully cleaned following completion of the purification of the new gallium. Tests in May of 1991 indicated that the levels of residual $^{68}$Ge and radon were well below the signal predicted by the SSM. Separate extractions of the new and old gallium were carried out in June, July, and August. Beginning in September 1991, combined extractions were begun on all 57 tons. Very preliminary analysis of the first few runs is consistent with the earlier data, namely there is no significant rate of $^{71}$Ge.

Intensive work is under way to reduce noise pulsing and backgrounds in the L peak. This effort includes rebuilding of one of the counting systems with new preamplifiers, extensive filtering of all power lines, installation of a Faraday cage around the counting systems, improved passive shielding, studies of noise reduction using data from a 1-GHz transient digitizer, and installation of an additional stage of cryogenic distillation in the gas synthesis before filling of the counters. It is hoped that this will allow us to count the L peak in the near future, which would almost double our counting efficiency.

7. Conclusions

Let us compare the data obtained with the predictions of the SSM. Different SSMs predict that the total expected capture rate in $^{71}$Ga is in the range$^{1-2}$ of 125 to 132, with the dominant contribution (71 SNU) coming from the p-p neutrinos. The minimum expected rate in a Ga experiment, assuming
only that the Sun is presently generating nuclear energy at the rate at which it is radiating energy, is 79 SNU\textsuperscript{23}. Observation of significantly less than 79 SNU in a gallium experiment is difficult to explain without invoking new neutrino properties.

The first measurements from a gallium solar neutrino experiment have observed fewer $^{71}\text{Ge}$ atoms than predicted by the SSM. If the extraction efficiency for $^{71}\text{Ge}$ atoms produced by solar neutrinos is the same as for natural Ge carrier, the first measurements indicate that the flux may be less than that expected from p-p neutrinos alone. Thus, the solar neutrino problem may also apply to the low-energy p-p neutrinos, indicating the existence of new neutrino properties.

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9. References