THE BAKSAN GALLIUM SOLAR NEUTRINO EXPERIMENT

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ABSTRACT. A radiochemical ^{71}Ga-^{71}Ge experiment to determine the integral flux of neutrinos from the sun has been constructed at the Baksan Neutrino Observatory in the USSR. Measurements have begun with 30 tonnes of gallium. The experiment is being expanded with the addition of another 30 tonnes. The motivation, experimental procedures, and present status of this experiment are presented.

The inverse beta-decay of ^{37}Cl was proposed to measure the solar neutrino flux more than forty years ago [1,2]. Such a measurement was initiated in the 1960's in the Homestake gold mine, and the first result of the ^{37}Cl-^{37}Ar experiment was that the solar neutrino capture rate was less than 3 SNU [3] (1 SNU = 10^{-36} captures per target atom per second). This value disagreed with the then accepted theoretical prediction of 8-29 SNU [4]. Since then there have been many new measurements of the parameters used in calculating the solar neutrino flux, such as the nuclear reaction cross sections [5] and the composition of the solar surface [6]. These new measurements were incorporated in the Bahcall and Ulrich Standard Solar Model (SSM), which predicts 7.9 ± 2.6 (3σ) SNU for ^{37}Cl [7]. Another solar model calculation of Turck-Chieze et al. that uses somewhat different input data and physical assumptions gives 5.8 ± 1.3 SNU [8].

The overall average capture rate in ^{37}Cl measured at the Homestake installation from 1972-1988 was 2.2 ± 0.3 (1σ) SNU [9]. This disagreement between the theoretical calculation and experimental

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measurements is often referred to as the solar neutrino problem (SNP). There are some time intervals when the average capture rate in the ^{37}Cl experiment has been different from this overall average. For example, from 1987-mid 1988 the rate was 4.2 ± 0.8 SNU [10]. Measurements during this same time period by the KAMIOKANDE II water Cherenkov detector [11] gave $0.46 \pm 0.13(stat.) \pm 0.08(syst.)$ of the value predicted by the SSM, consistent with the ^{37}Cl measurement.

The ^{37}Cl and KAMIOKANDE II experiments are sensitive primarily to the high energy ^{8}B solar neutrinos, which constitute less than 10^{-4} of the total estimated solar neutrino flux. The neutrinos produced in proton-proton fusion in the Sun (p-p neutrinos), which make up the bulk of this flux, are far below the energy thresholds of these experiments. The p-p neutrino production rate in the Sun is fundamentally linked to the observed solar luminosity, and is relatively insensitive to alterations in the Solar Model. The need for an experiment capable of detecting these low-energy neutrinos (endpoint energy 420 keV) has been apparent for many years. Such an experiment may be able to differentiate between solar model and particle physics explanations for the solar neutrino problem. This paper describes and gives the present status of the radiochemical solar neutrino experiment to detect the p-p neutrinos based on the ^{71}Ga-^{71}Ge transition that is being performed by a Soviet-American collaboration at the Baksan Neutrino Observatory of the Institute for Nuclear Research of the Academy of Sciences of the USSR.

1. Scientific Purposes

1.1 Solar Neutrinos

According to the Bahcall and Ulrich SSM [7] the dominant contribution to the total expected capture rate with ^{71}Ga (132 ± 20 SNU) arises from p-p neutrinos (71 SNU). Contributions made by ^{7}Be (34 SNU) and ^{8}B (14 SNU) neutrinos are also important. If the observed capture rate is significantly below 70 SNU, it would be very difficult to explain without invoking some phenomenon like Mikheyev-Smirnov-Wolfenstein (MSW) oscillations, a clear signal of physics beyond the standard model [12-16].

A capture rate in the vicinity of 70 SNU, i.e. the value predicted by the SSM for the p-p neutrinos alone, would most probably imply that both the ^{7}Be and ^{8}B neutrinos are suppressed. Such a suppression might be accounted for by new solar physics as well as by new weak interaction physics. For example, it could be understood if the solar interior were approximately $2 \cdot 10^{6}$ K cooler than predicted by the SSM or it could be the result of the MSW effect [13] with the parameters of oscillation $\delta m^2 = 10^{-5}$ eV^2 and $|\sin 2\theta| \geq 8 \cdot 10^{-2}$ (assuming predominant mixing of two neutrino states).

If the observed capture rate is greater than 70 SNU, the situation will be more complicated. If the rate is approximately 120 SNU, it will be most natural to attribute this to the combined effect of all neutrinos except those produced in the decay of ^{8}B. This could be understood either within the framework of the "cool Sun" model with the solar interior temperature approximately 10^{6} K cooler than predicted by

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the SSM, or as a result of the MSW effect with $\delta m^2 = 10^{-4}$ eV^2, $|\sin 2\theta| \geq 8 \cdot 10^{-2}$.

1.2 Supernova Neutrinos

The gallium neutrino experiment can also detect neutrinos from a supernova explosion. They could be distinguished from solar neutrinos by production of an appreciable number of ^{69}Ge atoms. According to recent experimental measurements of the isobaric analogue state in ^{69}Ge [17], 10-20 ^{69}Ge atoms are estimated to be produced in 60 tonnes of gallium by a supernova explosion at the center of the Galaxy.

1.3 Test of Electric Charge Conservation

The Ga-Ge experiment provides a unique opportunity to test electric charge conservation [18-21]. The mass of the nucleus ^{71}Ga is greater than that of the nucleus ^{71}Ge, but the conservation of electric charge prohibits the decay $^{71}\text{Ga} \rightarrow {}^{71}\text{Ge}$ + any neutral particle(s) (e.g., $\nu + \tilde{\nu}$, gamma-quanta, etc.). The best current limit on electric charge conservation with respect to nucleon decay was set with 300 kg of gallium in 1980 [22].

If the measured flux from the Sun turns out to be consistent with that predicted by the SSM minus ^{8}B neutrinos, one will be able to set the following limit:

$$T_{1/2}(^{71}\text{Ga} \rightarrow {}^{71}\text{Ge} + \text{any neutral particle(s)}) \geq 2 \cdot 10^{26} \text{ yr}.$$

Under some reasonable assumptions this may be expressed in terms of the element-independent ratio of branching probabilities for the elementary neutron decay [20]:

$$\epsilon^2 = \frac{\Gamma(n \rightarrow p + \nu + \tilde{\nu})}{\Gamma(n \rightarrow p + e^- + \tilde{\nu}_e)} \leq 10^{-26},$$

which would be a three order of magnitude improvement over the current result [22].

1.4 Test of ν_e - $\tilde{\nu}_e$ Non-Identity

One more result which may be obtained with the Ga-Ge experimental facilities is a new limit on ν_e - $\tilde{\nu}_e$ non-identity. This experiment would be analogous to a planned calibration experiment [23,24]. Experiments of this kind have been performed at a nuclear reactor [25] and at an accelerator [26]. One proposal is to place a gallium target into the antineutrino flux from a reactor [27]. An alternative experiment is to irradiate the gallium target with antineutrinos from a β^--active isotope with a halflife of a few tens of days. Possible sources [28] are ^{32}P, ^{170}Tm, and ^{90}Y (produced from the readily-available ^{90}Sr). If 10 tonnes of metallic gallium in the actual geometry of the gallium-germanium experiment is used [23] and if it is

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assumed that the solar neutrino flux (the main source of background) corresponds to the SSM [7,11], the following limit might be set on the ratio $\alpha^2 = \sigma_{exp}/\sigma_{theor}$ ($\nu_e = \tilde{\nu}_e$) ≤ 0.006 with a ^{32}P source with an initial activity of 1 MCi. Such a result would be almost an order of magnitude improvement over the best current limit derived from [26] (see [27]).

2. History

The reaction $\nu_e + {}^{71}Ga \rightarrow e^- + {}^{71}Ge$ as a tool for detecting solar neutrinos was first proposed by V. A. Kuzmin [29] in 1964. The experiment seemed impractical at that time because of the very low worldwide production and the high cost of gallium. There also was no method developed for extracting and counting single ^{71}Ge atoms from the gallium target. The desirability of the Ga-Ge experiment became widely recognized in the 1970's, and intensive work began [30-36] on the chemical extraction of Ge from Ga, the counting of ^{71}Ge decays, and the production of the necessary amount of gallium in the Soviet Union.

Techniques of extracting minuscule amounts of Ge from Ga, essential for a gallium-germanium neutrino experiment, were developed for two detector materials: (1) gallium in the form of a concentrated hydrochloric acid solution [33] and (2) molten metallic gallium [30]. The experiment at Baksan uses a metallic gallium target. The most important advantages of the metal are reduced sensitivity to both internal and external backgrounds and a smaller volume. The smaller target volume results in a greater capture rate for electron neutrinos from a ^{51}Cr calibration source placed inside the gallium-containing vessel. The major disadvantages of the gallium metal target are the need for additional steps of chemical processing and somewhat greater quantities of fresh reagents for the extraction procedure.

3. Scheme of the Experiment

Figure 1 presents the overall scheme of the experiment.

3.1 Chemistry

The gallium is contained in chemical reactors that are lined with teflon and have an internal volume of 2 m^3. The reactors are provided with stirrers and with heaters that maintain the temperature just above the gallium melting point (29.8 C). Each reactor holds about 7 tonnes of gallium.

At the beginning of each run, a known amount of Ge carrier is added to the Ga in the form of a solid Ga-Ge alloy. The reactor contents are stirred so as to thoroughly disperse the carrier throughout the Ga metal. After a suitable exposure interval (3-6 weeks), the Ge carrier and any ^{71}Ge atoms that have been produced by neutrino capture are chemically extracted from the gallium.

It has been shown that contacting 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 thus begins

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by adding to each reactor an extraction solution containing 1 kg of HCl, 5.2 kg of H_2O_2, and 68.8 kg of H_2O. The total volume of the extraction solution 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_2O are purified before use. So as to minimize heating of the Ga, the HCl solution is cooled to -15 C and the water to 4 C. The mixture is intensively stirred and the Ga metal turns into a fine emulsion. The Ge dissolved in the Ga migrates to the surface of the emulsion droplets. In 7-9 min. the H_2O_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.

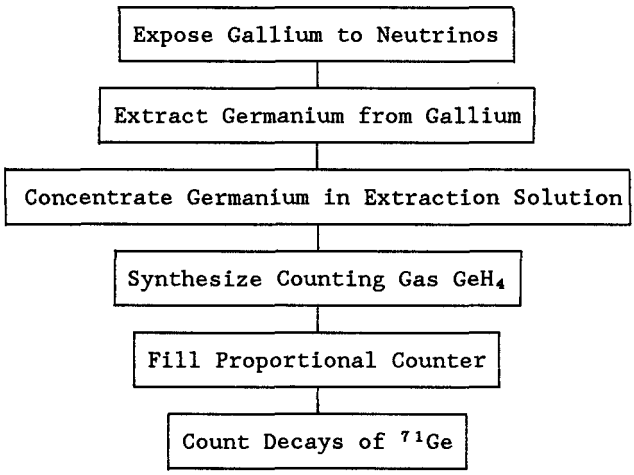

Figure 1: Scheme of the Experiment

All of the extracts from the separate reactors are combined and the Ge is then concentrated by vacuum distillation in 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/hour for 1.5 hours) sweeps the Ge as $GeCl_4$ from this acid solution into a volume of 1.0 l of H_2O. 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_2O. 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,

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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 gas GeH_4. The synthesis reaction that is used is optimized at a pH of 8-9, so NaOH is added to adjust the pH to this range. The resulting solution, whose volume is only 100 ml, is put into a small reactor flask in a sealed helium flow system and 50 ml of a 0.02N NaOH solution that contains about 2 g of $NaBH_4$ is added. GeH_4 is produced when this mixture is heated to about 70 C. The helium flow sweeps the GeH_4 into a gas chromatography system where it is purified. A measured quantity of xenon is added, and this mixture is put into a sealed proportional counter with a volume of about 0.75 cm^3. The GeH_4 sample is then counted for 1-6 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.

3.2 Counting

^{71}Ge decays with an 11.4 day half life. The decay occurs solely by electron capture to the ground state of ^{71}Ga. The probabilities of K, L, and M-capture are 88%, 10.3%, and 1.7%, respectively. The only way to register such a 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 10.26 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 similar to that used in the Cl-Ar experiment. In these counters it is not practical to separate the weak signal arising from the 0.12 keV events from background and the X-rays almost always escape. If the electrons are detected with 100% efficiency, then the counter energy spectrum consists of events at 10.4 keV (41.5% of all decays) and 1.2 keV (51.5% of all decays).

The SSM predicts a production rate of 1.2 atoms of ^{71}Ge per day in 30 tonnes of Ga. At the end of a 4 week exposure period, 16 ^{71}Ge atoms will be present on the average. If it is assumed that there is a one day delay between the end of exposure and the beginning of counting, that Ge is extracted from the Ga and transferred to the proportional counter with 75% efficiency, and that the counting efficiency is 35%, then the mean number of detected ^{71}Ge atoms in each run is only 3.9.

The major source of background in the counters is local radioactivity. The counters are thus made from materials especially selected to have a low content of Ra, Th, and U (such as synthesized quartz and zone-refined iron) and are contained in large passive shields made from copper, lead, and tungsten. The counting is conducted deep underground where the nuclear component of the cosmic ray flux is negligible and the muon flux is very small. To further reject and to characterize background events, some counting channels have a NaI

detector around the proportional counter that is used in coincidence/anticoincidence.

Background radioactivity primarily produces fast electrons in the counter. In contrast to the localized ionization produced by the Auger electrons from ^{71}Ge decay, these fast electrons give an extended ionization as they traverse the counter body. Since the risetime of the induced pulse increases as the radial extent of the ionization distance, it is possible to use pulse shape discrimination to separate the ^{71}Ge decays from background. This can be achieved by measuring the full pulse waveform for a few hundred nanoseconds or by differentiating the pulse with a time constant of about 10 nanoseconds and measuring the resulting amplitude (ADP method [37]).

Good rejection of background events occurs if the counting mixture contains 10% GeH_4 and 90% Xe at atmospheric pressure. This gas mixture gives a resolution of 18-21% at 5.9 keV. Some of the 9.2 keV X-rays are absorbed in this gas mixture and the measured total counting efficiency in a 2 FWHM window around 10.4 keV is 35%. This efficiency includes geometrical effects inside the counter and excludes events whose risetime is outside a 95% acceptance window.

3.3 ^{71}Ge Background

The main source of ^{71}Ge in the reactors other than from solar neutrinos is from protons arising as secondary particles produced by (1) external neutrons, (2) internal radioactivity, and (3) cosmic-ray muons. These protons can initiate the reaction ^{71}Ga(p,n)^{71}Ge. Extensive work has gone into measurements and calculations of these three background channels.

(1) Since the (n,p) cross sections on the Ga isotopes are small and the laboratory has been lined with low-background concrete, the fast neutron background is not significant. A calculation [38] has shown that the external neutron background in 30 tonnes of Ga metal produces no more than 0.01 atoms of ^{71}Ge per day.

(2) The background from internal radioactivity is mainly determined by the concentrations of U, Th, and Ra in the gallium. Measurements [39] of these concentrations, combined with measured yields of ^{71}Ge from alpha particles [33], indicate that less than 0.01 atoms of ^{71}Ge will be produced per day in 30 tonnes of Ga metal.

(3) Based on the measured muon flux in the laboratory, the production rates of the germanium isotopes from cosmic-ray muons have been calculated [40] to be 0.005 ^{71}Ge, 0.013 ^{69}Ge, and 0.009 ^{68}Ge atoms per day in 30 tonnes of Ga metal.

A valuable feature of the gallium experiment is that the ratios of the production rates for the isotopes ^{68}Ge, ^{69}Ge, and ^{71}Ge that arise from these background processes differ sharply from the corresponding ratios for production by solar neutrinos. This fact can be used to identify the source of the observed signal in the Ga-Ge detector. These background processes are all sufficiently small that the Ga experiment has the capability of observing a significant depression of the solar neutrino flux below the SSM predicted value.

Since these sources of ^{71}Ge background have all been made small,

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the major difficulty for the experiment that they pose is the need to remove from the gallium the large quantities of long-lived ^{68}Ge (half life = 271 days) that was produced by cosmic rays while the gallium was on the surface. ^{68}Ge decays only by electron capture, so its decays cannot be differentiated from those of ^{71}Ge. The subsequent decay of ^{68}Ga (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}Ga decay gives an energy spectrum with a broad peak whose maximum is at about 1.0 keV. These ^{68}Ga decays can generally be identified by rise-time analysis of the counter pulse and by detecting a coincidence pulse in the surrounding NaI crystal.

Another type of background that arises only during counting can come from tritium in the counting gas GeH_4. So as to eliminate this source of counter background, special methods for synthesizing $NaBH_4$ have been developed [41] that use starting ingredients that can be selected to have a low tritium content.

4. Present Experiment Status

The gallium-germanium neutrino experiment is situated in an underground laboratory specially built in the Baksan Valley of the Northern Caucasus, USSR. 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 muon flux has been measured to be $(2.4 \pm 0.3)\cdot 10^{-9}$ $cm^{-2}sec^{-1}$. To reduce the background from fast neutrons, the laboratory has been fully lined with 60 cm of low-radioactivity concrete and a 6 mm inner steel shell. Reactor filling began in April 1988 and the experiment now contains 30 tonnes of gallium in four reactors. Each reactor has undergone at least 15 extractions to remove the germanium isotopes that were produced by cosmic-ray interactions while the Ga was on the surface.

The efficiency of extraction of germanium from the reactors is measured at several stages of the extraction procedure. When the efficiency is to be determined, inert Ge carrier (approximately 120 micrograms alloyed into 50 g. of Ga) is added to each reactor. As the process of germanium concentration is carried out, several samples are routinely taken and the Ge content in each is determined by atomic absorption analysis (AAS). 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 H_2O and after the back-extraction from CCl_4 into H_2O. A final determination of the quantity of germanium is made by measuring the volume of synthesized GeH_4. Table 1 gives typical results of germanium content in some of these samples. 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 Ge in the extraction solutions is then measured by AAS. The error on the quantity of Ge in the carrier is estimated to be $\pm$ 5%.

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Table 1: Measurements of Germanium Yields During Extraction Process

Results are corrected for losses due to the samples taken for analysis and are expressed in micrograms of Ge.

Germanium Carrier Initially Added to Four Reactors	468 ± 23
Germanium in Extraction Solution before GeH_4 Synthesis	366 ± 12
Germanium in Synthesized GeH_4	342 ± 10

The overall extraction efficiency from the Ge added to the reactors to the synthesized GeH_4 is approximately 75%. The standard procedure is to conduct three extractions in series within a period of 5 days without adding additional carrier to the reactors. The GeH_4 samples from each of these three extractions are usually counted separately.

Two low-background counting systems, each with four channels, have been used to count the GeH_4 samples. Four counting channels have a NaI crystal for background suppression and recognition of ^{68}Ge-decay events. Both of these systems use the ADP-method for pulse shape discrimination. The energy, ADP, time, and NaI energy are recorded for each event.

The total background rate of selected counters filled with 90% Xe, 10% GeH_4 has been measured in the energy interval of 0.7-13.0 keV to be approximately 1.5 events per day. In the region of the Ge K-peak energy, the counter background is 1 event per month.

Counting of the germanium samples began during the extraction of the germanium isotopes produced by cosmic-ray interactions. After the gallium had been stored underground for 3 months, the initial activity of the Ge in the 30 tonnes of Ga was 7700 events per day in the energy region of the Ge K-peak. For recent runs, the activity in the Ge K-peak (35% counting efficiency) is less than 1 count per day.

Some residual radioactivity is still present that produces events in the energy range of 1-15 keV. These events are predominantly below 6 keV; they have both slow and fast risetime components with some events in coincidence with a NaI detector. In recent extractions this activity does not appear to be decreasing at the same rate as in the earlier extractions. Many measurements of the activity obtained in various isolated steps of the extraction procedure have been conducted, but the statistics available are quite low and the source of this activity has not yet been definitely identified.

The counting rate after the latest extractions enables one to set a limit on the half life of ^{71}Ga for non-charge conserving decay of $T_{1/2}$ ($^{71}Ga \rightarrow {}^{71}Ge + \ldots$) $> 3 \cdot 10^{25}$ years. This is a two order of magnitude improvement over the best previously existing limit [22], and gives the value $\epsilon^2 < 7 \cdot 10^{-26}$.

5. Future Plans

Monthly extractions from the 30 tonnes of gallium will continue. Simultaneous with this work, further experiments to understand the source of the remaining background will be conducted and the detector

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will be expanded to contain 60 tonnes of gallium. The additional 30 tonnes is now stored underground and will be put into reactors in the next few months. Almost all of the glassware for the 60-tonne extraction system is installed and tests of the full-scale extraction system are now being conducted. At the background levels presently achieved, the full 60-tonne detector should yield a statistical accuracy of better than 25% after one year of operation if the production rate is 132 SNU.

The number of channels in the present counting systems will very soon be increased to 12. An additional 12 counting channels with full pulse shape digitization and storage are expected to be operational this summer. Significant quantities of ^{72}Ge, ^{73}Ge, and ^{76}Ge are available that will be used sequentially for the Ge carrier. Mass spectrometric analysis of the isotopes in the germanium extracted from the reactors should yield an improved understanding of the chemical process.

A calibration of the gallium detector is planned using a reactor produced ^{51}Cr neutrino source. It is expected that an activity of 0.8 MCi will be obtained by irradiating about 200 g of enriched chromium (86% ^{50}Cr) in Soviet reactor SM-2 (thermal flux = $3.2 \cdot 10^{15}$ neutrons/cm^2-sec [42,43]). Approximately 400 decays from the ^{71}Ge atoms produced by this source are expected to be detected, yielding a statistical accuracy in the calibration of 5%.

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