Reports

Limits on Sensitivity of Large Silicon Bolometers for Solar Neutrino Detection

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Estimates are given for ultimate limits on background in proposed direct-counting measurements of neutrino scattering from large silicon crystals. Methods of background reduction are discussed. In the best case, the limiting backgrounds due to activities from cosmic-ray spallation of silicon would be less than the expected true event rate for reactor neutrino measurements of coherent neutral-current scattering from silicon nuclei. Considerable reduction of the estimated high-energy backgrounds would be required for a good signal-to-noise ratio in solar neutrino detection.

RECENT TECHNOLOGICAL ADvances have enabled several groups to detect single nuclear particles by bolometry (*1–3*). For example, x-rays or alpha particles have been detected with silicon (*1*) or diamond (*2*) bolometers, respectively, each weighing a tiny fraction of a gram. The use of very large bolometers for detecting neutrinos from the sun and from other sources has been discussed (*4, 5*), and several groups are developing bolometric (thermal) detectors with large mass (*6, 7*). Our group at Stanford is developing detectors that detect energy deposited by radiation before thermalization, in the form of ballistic phonons (*8, 9*). Silicon offers advantages as a detector material, having a good balance between large neutral-current scattering cross section (proportional to the square of the neutron number) and large energy transfer (inversely proportional to atomic number). The highly developed technology of silicon device manufacture and silicon purification may be used to advantage in detector development.

Multikilogram silicon single crystals at a temperature of a few hundred millikelvins would be instrumented to detect the few kiloelectron volts deposited as phonons by nuclei or electrons recoiling from single elastic scatterings of neutrinos. This scheme is attractive because of the large cross sections for coherent elastic scattering (*10*) of neutrinos and the potential for good temporal, spatial, and energy resolution (*8, 9*). Such detectors would have the unique capability of detecting the very small energy transfers due to the elastic scatters, and might also be useful for high-resolution detection of other nuclear particles (*8*) and for detection of certain classes of astrophysical dark matter particles (*11*).

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Table 1 shows the event rates per unit detector mass expected from various neutrino sources, including scattering from electrons and nuclei. With these low specific rates, it is essential to carefully consider the possible sources of background. In this report, an attempt is made to estimate the expected counting rates due to radioisotopes and cosmic-ray interactions in the silicon. The calculations indicate that cosmic-ray spallation–produced nuclides will not interfere with reactor-neutrino measurements. Available data from double-beta decay germanium detector experiments are also used to compute backgrounds in a large neutrino detector, the results again indicating the feasibility of a reactor experiment.

The scaled high-energy background from the germanium experiments is much larger than the expected solar neutrino signal, as are the calculated rates from cosmogenic ^{3}H and ^{22}Na in silicon. However, since the source of the background in the germanium experiments is not well understood, the mass scaling used may be overly pessimistic, and rejection may be possible by event topology discrimination. The cosmogenic nuclides may also be removable or their production prevented by suitable precautions.

The main elements occurring in purified silicon are silicon, carbon, oxygen, boron, and phosphorus (*12*). The long-lived radioisotopes ^{14}C and ^{32}Si are continually produced by the actions of cosmic rays on atmospheric nitrogen, oxygen, and argon. Material containing these elements that is in equilibrium with the atmospheric isotope distribution (a living organism, for example) will contain the radioisotopes at known abundances. Radiocarbon dating, for example, is based on the abundance reduction due to radioactive decay when material is cut off from exchange with the atmospheric reservoir.

Table 1. Estimated interaction rates per unit detector mass for various neutrino sources (1-keV minimum energy deposit).

Source	Neutrino flux (cm^{-2} sec^{-1})	Interaction rate (kg^{-1} day^{-1})
Reactor	10^{13}	100
Sun	6×10^{10}	1.5×10^{-3}
Submarine*	10^{7}	10^{-4}
Fission explosion†	6×10^{14} cm^{-2} (per blast)	10^{-2} (per blast)

*Ten-megawatt (thermal) reactor at 1 km. †One hundred kilotons at 1 km, 50% fission yield.

Silicon derived from deep underground silicate rock is typical "inorganic" matter (that is, either never in equilibrium with the atmospheric reservoir or isolated from it for a very long time). From accelerator mass spectrometry of inorganic carbons one can estimate minimum effective isolation times of about 50,000 years (*13*). This limit is probably a lower limit, since the measurements are not likely to correspond to actual residual ^{14}C content, but rather to backgrounds intrinsic to the detectors and contamination by organic materials.

With an effective isolation time of 50,000 years, the measured ^{32}Si activity of "recent" biogenic silica (3×10^{4} disintegrations per day per kilogram of silicon) (*14*), and the decay half-life of ^{32}Si (300 years), one predicts a totally negligible decay rate from preexisting ^{32}Si in inorganic silica (10^{-51} disintegrations per kilogram per year). A similar calculation for ^{14}C in silicon based on the relevant organic abundance (*13*) (1.2×10^{-12}) and half-life (5700 years) gives about 3.6×10^{-4} disintegrations per kilogram per day, which is also small.

An a priori estimate of the cosmogenic ^{32}Si activity expected in selected silicon can also be made. Consider a mine at 1000 m water-equivalent (mWE) depth, with silicate rock containing 1% by weight potassium impurity. [Commercially exploited silicate sources with much better characteristics than these are found in California (*15*).] The ^{32}Si-production cross section by spallation of potassium may be taken as one-tenth of the ^{3}H-production cross section. With these assumptions, the saturation activity of cosmogenic ^{32}Si would be only about 10^{-6} disintegrations per kilogram per day, corresponding to roughly 3×10^{-21} atom percent ^{32}Si. Thus, ^{32}Si is not expected to contribute significantly to the backgrounds. The total activity of detector-grade silicon in the energy region of ^{32}Si decay events is experimentally known to be $\leqq 400$ disintegrations per kilogram per day. Concerning the remaining impurities, there are no radionuclides (oxygen, boron, or phosphorus) that are long-lived enough to contribute in this sort of calculation.

Nuclear interactions of the cosmic rays (mainly the nucleonic component) will produce radionuclides by spallation of the detector material (*16*) during any refining and fabrication processes carried out at the earth's surface. Consider first spallation of $Z > 14$ impurities in silicon. With 0.1% impurities [mines with 0.02% impurities exist (*15*)], and with tabulated spallation cross section fits (*17*), spallation of heavy impurities is calculated to produce for each day of exposure at sea level a few times 10^{-6} disintegrations per day of ^{32}Si and heavier nuclides per kilogram of SiO_2, which is negligible.

Spallation of the silicon itself will produce light radionuclides. Suppose silicon is exposed at sea level for 3 months during refining, followed by 1 year of "cooling off" deep underground. Again with published spallation cross sections and fits (*17*) and cosmic-ray intensities (*16*), the principal activities remaining after cool-down turn out to be ^{3}H, ^{22}Na, and ^{7}Be at the levels shown in Table 2. These activities could be reduced by carrying out the refining and detector fabrication underground. Even a modest overburden of 25 mWE would reduce the spallation rate by a factor of about 100 (*18*).

It may also be possible to eliminate the remaining activity. It is known that some impurities, including hydrogen and sodium, diffuse very rapidly at high temperatures in silicon (*19*), suggesting that ^{3}H and ^{22}Na may be quantitatively removed by isotopic exchange during annealing in nonradioactive hydrogen or sodium gas. The ^{7}Be would remain as a convenient internal standard (480-keV γ-ray) at a tolerable rate. This diffusion exchange method has been used as a standard radiochemical method for determination of ^{3}H in materials such as iron and aluminum (*20*).

Events from the passage of cosmic-ray muons (about 10 per kilogram per day in a neutrino observatory at 1000 mWE depth) can presumably be externally vetoed with 99% or better efficiency. Neutrons from muon capture can produce counts that are considerably delayed with respect to the incident muon and therefore cannot be electronically vetoed. Estimates of this contribution at depth have been performed (*21*) and are extremely small.

Direct information on residual activity in hyperpure germanium is available from germanium double-beta decay experiments (*22–25*). In these experiments the decays

$$^{76}Ge \rightarrow {}^{76}Se + 2e \quad (1)$$

and

$$^{76}Ge \rightarrow {}^{76}Se + 2e + 2\nu \quad (2)$$

are sought within several-hundred-gram germanium diodes in special low-background cryostats, generally in underground laboratories. Of course, the sources, purification, and nucleonics of germanium are rather different from those for silicon, so the germanium double-beta decay results are only indicative of possible limits to be encountered with silicon.

Table 2. Cosmogenic activities present in pure inorganic silicon after 3 months of cosmic-ray exposure at sea level followed by 1 year of decay deep underground.

Nuclide	Specific activity (disintegrations per kilogram per day)
^{3}H	3.5
^{22}Na	0.32
^{7}Be	0.07*
^{14}C	1.1×10^{-4}
^{10}Be	2.1×10^{-6}
^{26}Al	2.9×10^{-7}

*6.4 per kilogram per day before cool-down.

The energy region of interest for decay process 1 lies at 2.04 MeV, so most of the efforts over the longer term have gone into reduction of "high-energy" backgrounds. Recent interest in dark matter detection has led to efforts at background reduction in the energy region near a few kiloelectron volts by at least one group (*26*).

The lowest published rates so far obtained in germanium double-beta decay experiments are a continuum with about 5 counts per day per kilogram per kiloelectron volt near 10 to 20 keV, with superimposed peaks due to gallium x-rays [from $^{70}Ge(n, \gamma)^{71}Ge$, followed by electron capture decay to ^{71}Ga], at about 20 counts per day. Uranium and thorium alpha and gamma peaks were observed at a total rate of about 1 count per kilogram per day, above a continuous background of about 0.01 count per day per kilogram per keV in the "high-energy" region near 2 MeV (*22, 23*).

Most workers believe the low-energy continuum arises from (n, γ) reactions or radioactive decays in the surrounding shielding. The neutron flux from fission in the rock surrounding an underground lab depends upon the geology, but at Gotthard and Sudbury it is a few times 10^{-5} cm^{-2} sec^{-1}. With no shielding, these neutrons would scatter at least once, or be captured in a silicon detector, leading to a continuum with a total rate of roughly 1000 captures per kilogram per day. Appropriate shielding would reduce the neutron rate by at least an order of magnitude to a level consistent with the germanium results quoted.

To estimate the rate in a 100-kg silicon detector from the double-beta decay experiment data, the continuum rates are scaled with detector volume, resulting in a scaled rate of 500 counts per keV per 100 kg per day in the low-energy continuum and 1 count per keV per 100 kg per day for the high-energy region. This gives an overestimate because the silicon detectors, unlike germanium diodes, are expected to have spatial resolution. Events with the poorly localized energy deposition characteristic of neutron capture gamma rays—events originating in the shielding, cryostat material, or metallization layers of a silicon detector—could thus be greatly suppressed.

The low-energy continuum background affects mainly reactor measurements, since the bulk of the solar neutrinos have too little energy to be detected by coherent scattering (*4*). The scaled low-energy rate is just half the expected true event rate (Table 1).

The high-energy background would be important in solar neutrino detection, since the signal in this case is primarily due to electron-neutrino scattering with energy deposits of the order of a few megaelectron volts (*4*). The high-energy continuum background scaled from the double-beta decay experiments to the 100-kg silicon detector is much larger than the expected total event rate of about 0.15 total counts per day in a 100-kg detector. The rejection techniques mentioned above will reduce this background, but the rejection efficiency cannot be accurately estimated without better knowledge of the actual source and characteristics of the background to be rejected.

In summary, coherent neutral-current interactions of intense reactor neutrino sources give counting rates in excess of expected background limits. These interactions should be detectable with acoustic silicon detectors of modest size (a few kilograms). Direct counting of solar p-p neutrinos, interacting in silicon primarily by ν-e scattering, may be possible if several conditions are met. The silicon must be prepared from inorganic silica originating deep underground. Exposure to cosmic rays during refining must be held to a minimum. In addition, the high-energy background in such detectors must be made much lower than that obtained by scaling results from germanium double-beta decay date. This might be achieved either by improved radiopurity or by developing powerful electronic techniques for background rejection.

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Bacterial Methanogenesis and Growth from CO_2 with Elemental Iron as the Sole Source of Electrons

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Previous studies of anaerobic biocorrosion have suggested that microbial sulfur and phosphorus products as well as cathodic hydrogen consumption may accelerate anaerobic metal oxidation. Methanogenic bacteria, which normally use molecular hydrogen (H_2) and carbon dioxide (CO_2) to produce methane (CH_4) and which are major inhabitants of most anaerobic ecosystems, use either pure elemental iron (Fe^0) or iron in mild steel as a source of electrons in the reduction of CO_2 to CH_4. These bacteria use Fe^0 oxidation for energy generation and growth. The mechanism of Fe^0 oxidation is cathodic depolarization, in which electrons from Fe^0 and H^+ from water produce H_2, which is then released for use by the methanogens; thermodynamic calculations show that significant Fe^0 oxidation will not occur in the absence of H_2 consumption by the methanogens. The data suggest that methanogens can be significant contributors to the corrosion of iron-containing materials in anaerobic environments.

CORROSION OF METALS IS A SERIOUS economic problem. A significant amount of corrosion, particularly under anaerobic conditions, is thought to be mediated by microorganisms (*1–8*). The sulfate-reducing bacteria are widely regarded as the chief agents of biocorrosion in anaerobic environments. The corrosive activities of these organisms are due in part to their ability to form corrosive hydrogen sulfide (*1–3, 5–8*) and reduced phosphorus compounds (*4, 9*). In addition, these bacteria are thought to cause corrosion by cathodic depolarization (Fig. 1), a mechanism first proposed by von Wolzogen Kühr and van der Vlugt (*10*). According to this theory, the bacteria accelerate the anodic dissolution of metal by using hydrogen (formed from water-derived protons and cathode-derived electrons) through their hydrogenase enzymes. The reducing equivalents so generated are used in the dissimilatory reduction of sulfate to hydrogen sulfide:

$$8[H] + SO_4^{2-} + 2H^+ \rightarrow H_2S + 4H_2O \qquad \Delta G^{0'} = -152\ \text{kJ} \qquad (1)$$

[H] is used to describe hydrogen species of unknown structure, either as bound atomic H or H_2; $\Delta G^{0'}$ is the standard free energy change at *p*H 7. Recent work with hydrogenase-positive *Desulfovibrio* strains showed that iron was a partial source of the electrons involved in sulfate reduction; however, iron did not serve as a sole electron donor, since no sulfide was formed in the absence of lactate (*11*). Other workers have suggested that [H] is derived from a cathode of ferrous sulfide precipitated onto the metal surface (*7, 12*); this temporary cathode is thought to be continually regenerated by bacterial hydrogen removal (*7, 8*). Despite wide acceptance of the cathodic depolarization concept, there have been few unequivocal experimental demonstrations of its occurrence (*13*). There is no evidence that this phenomenon is coupled to microbial growth [except where electrically generated cathodic hydrogen supported the growth of sulfate-reducing bacteria (*14*)].

The requirement for a hydrogenase in the cathodic depolarization mechanism suggested to us that other hydrogen-using bacteria might perform cathodic depolarization in the presence of the proper electron acceptor and might couple the energy generated to bacterial growth. Non-sulfate-reducing bacteria that oxidize hydrogen would also be a cleaner experimental system for studying cathodic depolarization because problems with measuring growth, metal dissolution, and Fe^{2+} production in the presence of large amounts of precipitated sulfides could be avoided. In this report we show that methanogenic bacteria, which grow through anaerobic respiration, normally as in Eq. 2, are capable of growth and methane production with metallic iron as sole electron source:

$$4H_2 + CO_2 \rightarrow CH_4 + 2H_2O \qquad \Delta G^{0'} = -139\ \text{kJ} \qquad (2)$$

To our knowledge, this is the first demonstration of Fe^0 as an energy source for growth of any organism and thus represents a novel type of chemolithotrophic energy metabolism.

When mid-logarithmic stage cells of *Methanosarcina barkeri* were examined for their ability to oxidize Fe^0, H_2 was replaced

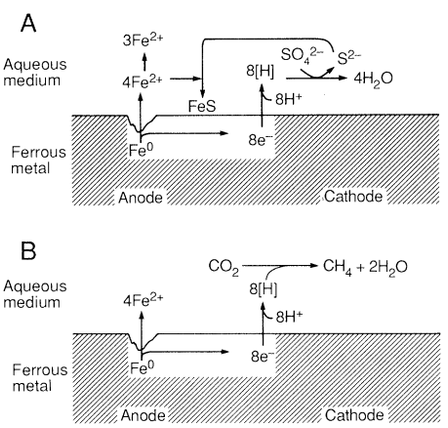

Fig. 1. (**A**) Schematic illustration of cathodic depolarization reactions proposed for corrosion of ferrous metals by sulfate-reducing bacteria according to the classical mechanism of von Wolzogen Kühr and van der Vlugt (*10*). The form of "hydrogen" is not specified. (**B**) A similar mechanism for cathodic depolarization by methane-producing bacteria.

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