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Experimental Search for Neutrinoless Double-\(\beta\) Decay of \(^{100}\)Mo

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Abstract

No evidence for the neutrinoless double-β decay of $^{100}$Mo has been found in a search using a segmented Si(Li) detector with source foils enriched to 97% $^{100}$Mo. After 0.2664 mole years of exposure, we report a 1 $\sigma$ lower limit on the half-life for the $J^p = 0^+ \rightarrow 0^+$ transition of $0.35 \times 10^{23}$ years.

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The existence of the neutrino was proposed over 60 years ago by Pauli, followed shortly thereafter by Fermi's theory of $\beta$ decay. Yet to this day there are fundamental unanswered questions concerning properties of the neutrino and of weak interactions in general. Neutrinoless double-$\beta$ decay directly addresses the question of lepton conservation in weak interactions as well as the related question of whether the neutrino has mass and, if it does, whether it is its own antiparticle. As such it is one of the most sensitive tests of physics beyond the standard model of elementary particles and for this reason has received much attention both theoretically and experimentally during the past decade. Despite considerable experimental progress, there has as yet been no convincing evidence for the existence of this decay mode reported in any element [1]. As a result, upper limits can be placed on the mass of a Majorana neutrino that are substantially better than those obtained by the more direct $\beta$ decay end-point method.

The isotope selected here as a neutrinoless double-$\beta$ decay candidate is $^{100}$Mo. The lepton-number violating neutrinoless decay mode, $^{100}\text{Mo} \rightarrow ^{100}\text{Ru} + 2e^-$, is forbidden for massless neutrinos or for Dirac neutrinos (which are distinct from their antiparticles). There have recently been three lower half-life limits reported for the neutrinoless decay of $^{100}$Mo, one by an earlier version [2] of this experiment with a limit of $4 \times 10^{21}$ years and two others [3,4] of 4.7 and $7 \times 10^{21}$ years. $^{100}$Mo is selected for its large energy release (which increases the phase space for neutrinoless double-$\beta$ decay), for its favorable double-$\beta$ decay matrix-element, and for the availability of several moles of the enriched isotope. Assuming comparable matrix elements, the product of phase space and Coulomb correction factor increase the $^{100}$Mo neutrinoless double-$\beta$ decay rate over other commonly used element isotopes for double-$\beta$ decay searches by factors ranging from 1.3 to 12.3 [5–8]. In addition, the 3.033 MeV energy release shared by the two electrons is energetically above most naturally occurring electron and photon background radiation.

The original apparatus has been described in detail elsewhere [9,10]. Data reported here were obtained from an upgraded detector array which contained 145 Si(Li) detectors, each 7.6 cm in diameter and 0.14 cm thick, arranged in two stacks. One stack with 74 detectors
was run without $^{100}\text{Mo}$ source foils for background studies. The second stack, containing 71 detectors, was loaded with 62 $^{100}\text{Mo}$ source foils placed in the gaps between detectors.

Titanium clips were used to provide more positive electrical contacts to the silicon detectors in the upgraded apparatus. Source foils consisted of mylar bags 6.0 cm in diameter, each containing approximately 1 gram of $^{100}\text{Mo}$. The very finely divided pure metallic $^{100}\text{Mo}$ powder had been chemically purified to reduce the Th and U levels to below 1 ppb. Each bag was rolled flat (to within 10%) before being inserted into a gap between detectors. The average thickness of the $^{100}\text{Mo}$ within each bag was 34.4 mg/cm$^2$, and the thin mylar walls (4.3$\mu$m) of the bags contributed a negligible amount of additional material. The entire detector array was enclosed in the radioactively clean titanium cryostat of the original apparatus and cooled to 120 K with liquid nitrogen.

Cosmic ray backgrounds are negligible at our underground site in the Consolidated Silver mine in Osburn, Idaho. The site has a rock overburden of 1220 m (3300 m.w.e.). Shielding of the detectors from natural radioactivity in the surrounding rock consisted of three concentric closed layers of appropriate materials to reduce neutrons and $\gamma$ rays. The neutrons were moderated by a 56 cm thick wax shield and were then captured in a 5.1 to 10.2 cm thick inner layer of 5% borated polyethylene. Any $\gamma$ rays penetrating the wax and polyethylene were substantially eliminated by absorption in the innermost layer of low activity lead, 25.4 cm thick. The interior of this lead shield was continuously flushed with nitrogen gas from liquid nitrogen boiloff to reduce airborne Rn daughter activity. All the structural parts of the apparatus within the shielding were constructed from materials selected by means of low level counting for low intrinsic radioactivity.

The upgraded detector included electronic latches on each channel to aid in identifying fast $\beta - \alpha$ decay sequences. The $\beta - \alpha$ time interval between a $^{212}\text{Bi}$ or $^{214}\text{Bi}$ beta decay and the decay of its Po daughter was measured by a 100 MHz “fast” clock that was started by each event and stopped by any second trigger occurring during the event read-out time of approximately 50ms. A 1 kHz “slow” clock measured time intervals between events and was used to detect longer-lived decay sequences. Lifetimes measured by these clocks allowed
identification of radio contaminants within the cryostat.

The absolute energy calibration of the experiment was checked periodically by placing a sealed $^{228}$Th source on the surface of the cryostat near the detectors and identifying the double escape peak at 1.593 Mev in each detector. In addition, before each data run, a digital to analog converter running under computer control was used to inject a series of pulses with precisely known amplitudes into the preamp of each detector to check the electronic gain and linearity of the readout system. Any drifts were recorded in software and corrected on a run by run basis. Throughout the experiment, the energy resolution of a typical detector was between 10 keV and 20 keV and its energy determination was stable between absolute source calibrations to within the detector's resolution.

The data from this experiment correspond to 3849.5 hours live-time with 60.63 gms of $^{100}$Mo in the detector stacks, giving an exposure of 0.2664 mole-yrs. There were 299,852 triggers above the nominal discriminator threshold of 320 keV. About 90% of the events had energy in only one detector. We ascribe the one-detector events with energies $2.0 < E < 5.5$ MeV to $\alpha$ decays of $^{210}$Po ($E_\alpha=5.3$ MeV), and events below 2 MeV to a combination of $^{210}$Po decays and single $\beta$ decays from various sources. The $^{210}$Po background probably came from $^{210}$Pb in solder used to make electrical contacts within the cryostat.

The following cuts, which were optimized by Monte Carlo simulations and studies of the real data, were made to select neutrinoless double-$\beta$ decay candidates and reject backgrounds: (a) To remove electronic noise, the energy in a detector was ignored if it was less than 110 keV. In addition, for multi-detector events any contribution to the event energy sum was ignored if it was less than $1/20$ the maximum contribution. (b) To remove $\alpha$ backgrounds, events with energy deposited in only one detector were rejected. (c) To minimize backgrounds from single $\beta$ decays, which are usually accompanied by $\gamma$ rays, only events with energy in two or three contiguous detectors were selected. In addition, the energy deposited in the middle detector of a three-detector event had to be greater than 450 keV (the minimum energy deposited by a single electron passing through a detector). (d) Events "tagged" by the fast clock were rejected, since they were usually $^{212}$Bi or $^{214}$Bi $\beta$ decays
followed by a fast $^{212}\text{Po}$ or $^{214}\text{Po}$ $\alpha$ decay. (e) Events with energy deposited in two detectors, where the energy ratio in the detectors was less than 1/3, were rejected. This reduced the number of untagged $^{212}\text{Bi}$ events in which the energy of the $\alpha$ from the subsequent fast decay of $^{212}\text{Po}$ was recorded in an ADC but the $\alpha$ arrived before the trigger logic had recovered and so did not stop the fast clock. (f) A possible neutrinoless double-$\beta$ decay candidate was rejected if there was energy deposited in a detector next to an inoperative detector, since the total energy of the event was then suspect.

After all these cuts there remain 8634 events in the region of the detectors occupied by the $^{100}\text{Mo}$. This corresponds to about 6% of the total number of events observed in this region of the detector. The energy spectrum for these double-$\beta$ decay candidate events, for energies above 1.8 MeV, is shown in Fig. 1 (728 events).

To obtain the detection efficiency for observing double-$\beta$ decay candidates and to estimate the spectral shapes of backgrounds, a Monte-Carlo event generating program was written to simulate double-$\beta$ decays and backgrounds within the geometry of our detector [11]. This program was a modification of the CERN program GEANT [12]. The generated events were processed through our standard analysis programs with all the cuts necessary to select double-$\beta$ decay candidates. The shape of the spectrum for double-$\beta$ decay candidates in the foils is shown on Fig. 1 and was fit to an analytical function ($\chi^2/df < 1.2$) which was used in fitting procedures to determine the double-$\beta$ decay half-life described below. The detection efficiency for the $0^+ \rightarrow 0^+$ transition is about 47% for all observed energies and 36% for energies above 2.5 MeV.

To investigate expected $\beta$ decay backgrounds, energy spectra for the $\beta$ decays of $^{214}\text{Bi}$, $^{212}\text{Bi}$, and $^{208}\text{Tl}$ were generated. The expected energy spectra for $\alpha$ decays were calculated using a simple Monte Carlo. In an attempt to estimate the size and probable sources of backgrounds, the experimental data and simulated data described above were examined carefully. Comparisons were made of event rates in stack 1 and stack 2, and also rates in stack 2 before and after the $^{100}\text{Mo}$ was inserted. An assessment of the $^{238}\text{U}$ and $^{232}\text{Th}$ activity within the cryostat was obtained by studying events tagged by the fast clock during event
read-out, or by observing rapid multi-α sequences in: (a) the β decay of \(^{212}\)Bi in the \(^{232}\)Th chain followed by an α decay of the daughter \(^{212}\)Po, which has a half-life of 0.3μsec; (b) similarly, the β decay of \(^{214}\)Bi in the \(^{238}\)U chain followed by an α decay of \(^{214}\)Po, which has a half-life of 164μsec; and, (c) the α-α-α rapid decay sequence, \(^{220}\)Rn → \(^{216}\)Po\((t_{1/2}=56\text{s})\) → \(^{212}\)Pb\((t_{1/2}=0.15\text{s})\) in the \(^{232}\)Th chain. These α sequences were observed as either two or three separate one-detector events in the same or adjacent detectors.

In the α decay sequences, if the contamination is within the \(^{100}\)Mo, the events have to be seen in the same detector. Because the \(^{100}\)Mo filled bag was thicker than the range of any of the α particles, back-to-back α particles would not give a sequence of events in adjacent detectors. We do, however, see sequences that were in adjacent detectors and can only be explained if most of the \(^{232}\)Th chain α decay sources were not in the \(^{100}\)Mo powder. A possible source of this contamination was the Ti clips which made electrical contact with the detectors and were constructed in such a way that they were visible to the detectors above and below them. By comparing the rates of fast clock tagged events for empty mylar bags and \(^{100}\)Mo filled mylar bags, we find that some, but not all, of the \(^{214}\)Bi β decays came from contamination in the mylar.

Estimated backgrounds above 2.5 MeV used in the final analysis were obtained from observed tagged \(^{212}\)Bi and \(^{214}\)Bi decays and Monte Carlo generated \(^{208}\)Tl decays. The \(^{212}\)Bi background was estimated to be flat for energies between 2.0 and 3.5 MeV and the \(^{208}\)Tl background above 2.0 MeV was fit to a straight line with a negative slope. Many analytical curves, including a straight line, a quadratic, and a parameterized exponential, were used to fit \(^{214}\)Bi background above 2.0 MeV. All the observed events above 2.5 MeV could be easily explained by various combinations of the estimated backgrounds.

Two independent analyses were carried out to determine the lifetime for \(0^+ \rightarrow 0^+\) neutrinoless double-β decay. In one, least squares fitting programs [13] were used to fit normalization parameters for the analytical expressions of the signal and the individual backgrounds to the 12 observed events above 2.5 MeV. In every case, the best fit to the data pushed the normalization parameter for the signal to a negative value or a zero lower limit as it at-
tempted to transform the signal's peak into a dip to fit the data, and the value of the signal's normalization parameter was insensitive both to the functions used to fit the backgrounds and to a wide range of values of background normalization parameters. Up to three artificial events could be introduced into the real data near the peak of the double-\(\beta\) decay spectrum before its normalization parameter became greater than zero in the fit. Based on this study, which finds no real signal events above 2.5 MeV, a value for \(N = \ln(1/(1 - 0.68)) = 1.14\) events can be obtained from Poisson statistics where \(N\) is defined as the mean of a Poisson distribution which will produce more than zero signal events 68% of the time in random observations. [14,15]

In the second analysis, our estimates and fits to the background and signal shapes above 2.5 MeV enabled us to use a numerical technique [16] to obtain a lifetime at the 68% confidence limit. Assuming a null signal and only background events above 2.5 MeV, which is the most likely result which fits the observed data, 1000 "virtual" data sets were randomly generated from Poisson statistics for several different background combinations, and refit to the same signal and background shapes. In 68% of the cases for each of the different data sets, the signal extracted ranged between 1.1 and 1.6 events depending on the relative amounts of \(^{212}\)Bi and \(^{214}\)Bi used as background.

The 1\(\sigma\) limit on the half-life for neutrinoless double beta decay can be calculated from the formula

\[
t_{1/2} \geq \frac{\ln(2) T \times N_a}{N}
\]

where \(T = 0.2664 \times 0.36 = 0.0959\) mole-years is the product of the livetime and detector efficiency, \(N_a\) is Avagadro's number, and \(N\) is the number of events. Using the estimate of 1.14 for \(N\), we obtain \(t_{1/2} > 0.35 \times 10^{23}\) years. This is nearly a factor of 9 larger than our previous result [2].

An estimate of 7.4 eV for the upper limit on the effective Majorana neutrino mass \(< m_\nu >\) for the 0.35 \(\times\) \(10^{23}\) year half-life limit obtained in this experiment can be calculated using the matrix elements of Engel [7]. It should be emphasized, however, that this estimate is both
model dependent and sensitive to the approximations used to calculate the matrix elements.

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FIGURES

FIG. 1. Energy spectrum of events after all cuts. The corresponding energy spectrum for the Monte Carlo generated $0^+ \rightarrow 0^+$ neutrinoless double-$\beta$ decays after the same cuts is shown as a solid line. This curve is normalized to 500 events for a suitable graphical representation.