Double Beta Decay of $^{100}\text{Mo}$ to Excited Final States

M. J. Hornish and L. De Braekeleer

The Department of Physics,
Duke University and Triangle Universities Nuclear Laboratory,
Durham, NC 27708-0308

A. S. Barabash and V. I. Umatov

Institute for Theoretical and Experimental Physics,
B. Cheremushkinskaya 25, 117259, Moscow, Russia

(Dated: April 20, 2018)

A systematic study of the inclusive $(0\nu + 2\nu)$ double beta ($\beta\beta$) decay of $^{100}\text{Mo}$ to various excited final states of $^{100}\text{Ru}$ was performed. Utilizing two large HPGe detectors operated in coincidence, a search for the subsequent deexcitation $\gamma\gamma$ cascades was conducted. A 1.05-kg sample of isotopically enriched (98.4%) $^{100}\text{Mo}$ was investigated for 455 days, yielding an unambiguous observation of the $\beta\beta$ decay of $^{100}\text{Mo}$ to the $0^+_1$ state (1130.3 keV) of $^{100}\text{Ru}$. This excited final state decays via the $0^+_1 \to 2^+_1 \to 0^+_0$, sequence (with $E_{\gamma 1} = 590.8$ keV and $E_{\gamma 2} = 539.5$ keV), and 22 such coincidence events were detected, with a continuous background estimated to be 2.5 events. This counting rate corresponds to a decay half-life for the $\beta\beta(0^+ \to 0^+_1)$ transition of $T^{(0\nu+2\nu)}_{1/2} = [6.0^{+1.9}_{-1.3}(\text{stat}) \pm 0.6(\text{syst})] \times 10^{20}$ years. Lower limits on decay half-lives were achieved for higher excited final states.

PACS numbers: 21.10.Tg, 23.40.-s, 27.60.+j

I. INTRODUCTION

A considerable escalation in the attention paid to the study of double beta ($\beta\beta$) decay is owed, in no small part, to the tremendous progress made by recent neutrino oscillation experiments. The remarkable success of solar and atmospheric neutrino experiments (see reviews [1, 2, 3]) represents a profound achievement and a veritable breakthrough in our understanding of the neutrino. More recently, the KamLAND reactor antineutrino result [4] and the latest SNO result [5] further reinforce the evidence for the oscillation of neutrinos. Indeed, there can no longer be any doubt that the neutrino is, in fact, a massive particle.

At the same time, however, for all of the progress that has been accomplished in measuring the mass differences ($\Delta m^2$) for neutrinos of unlike flavors, such oscillation experiments are inevitably and inherently insensitive to two additional important properties: (1) the neutrino’s absolute mass scale and (2) the Majorana-Dirac nature of the neutrino. The only present experimental method capable of attempting to simultaneously answer both of these questions is the search for the neutrinoless mode ($0\nu\beta\beta$) of double beta decay. Although this transition has not yet been observed beyond a reasonable doubt, the successful detection of this decay mode may also provide useful information about the neutrino mass hierarchy (normal, inverted, or quasi-degenerate) and lepton-sector CP violation via a measurement of the Majorana CP-violating phases (see discussions [6, 7, 8]).

The recent surge in interest in $\beta\beta$ decay has initiated the proposal of several large-mass experiments that will search for the neutrinoless mode (see review [3, 9, 10, 11, 12]). The interpretation of any future results from these experiments depends on the reliable estimation of nuclear matrix elements ($M^{0\nu}$) for the $0\nu\beta\beta$ transition. While the phase-space factor for this decay can be accurately calculated, any uncertainty in the corresponding nuclear matrix elements will adversely affect the ability to correctly extract Majorana neutrino mass information. Several theoretical models have been devised, including the well-known quasiparticle random phase approximation (QRPA), but a thorough analysis of the accuracy of the various models to properly calculate the nuclear matrix elements is difficult at present, although efforts are being made (see, e.g., [13, 14]).

One testing ground for our understanding of the $\beta\beta$-decay nuclear matrix elements is through the study of the ordinary allowed second-order weak decay, namely the two-neutrino double beta ($2\nu\beta\beta$) decay. In general, expanding our experimental knowledge of the $2\nu\beta\beta$ process for a variety of different nuclei will improve the overall comprehension of the nuclear part of double beta decay. Although the nuclear matrix elements for this mode ($M^{2\nu}$) are not identical to those of the neutrinoless mode, experimental measurements of $M^{2\nu}$ can be compared to theoretical model calculations as a check of the validity of the various theoretical schemes upon which the models rely.

Over the past decade and a half, significant progress has been made in successfully detecting the $2\nu\beta\beta$ de-
By searching for the subsequent $0^+_1 \rightarrow 2^+_1 \rightarrow 0^+_s$ decay cascade, with $E_{\gamma 1} = 590.79$ keV and $E_{\gamma 2} = 539.51$ keV (see Figure 1). One such experiment was performed by measuring enriched $^{100}$Mo samples ($\sim 1 \text{ kg}$) with a low-background HPGe detector. An analysis of the single $\gamma$-ray spectrum positively identified the $2\nu\beta\beta(0^+ \rightarrow 0^+_1)$ transition, and a deduced half-life of $T_{1/2} = (6.1^{+1.1}_{-1.1}) \times 10^{20}$ years was produced [24]. Afterwards, a second experiment using the same technique yielded a measured half-life for this transition of $T_{1/2} = (9.3^{+2.8}_{-1.7}) \times 10^{20}$ years, with a systematic error estimated to be $\sim 15\%$ [21]. By summing the single $\gamma$-ray spectrum of Ref. [24] to that of Ref. [21], one obtains a combined half-life of $T_{1/2} = (7.6^{+1.8}_{-1.7}) \times 10^{20}$ years, also with a systematic error of $15\%$ [21].

The preliminary results from the present experiment on $^{100}$Mo, with a quoted half-life of this $2\nu\beta\beta(0^+ \rightarrow 0^+_1)$ transition of $T_{1/2} = [5.9^{+1.7}_{-1.1}(\text{stat}) \pm 0.6(\text{syst})] \times 10^{20}$ years, were previously presented [23] and will be explored in greater detail in this Article.

II. EXPERIMENTAL DETAILS

By relying on single $\gamma$-ray searches, previous measurements of the $2\nu\beta\beta(0^+ \rightarrow 0^+_1)$ decay of $^{100}$Mo have involved the implementation of low-background detection systems, where detectors were made from low-radioactivity materials. These experiments were operated in underground laboratories, where the natural overhead shielding reduces cosmic-ray induced backgrounds. These experiments produce single $\gamma$-ray spectra exhibiting an excess of counts in the regions of interest near 539.5 keV and 590.8 keV, above a rather substantial background. In the two cases mentioned in Section I, the corresponding signal-to-background ratios were approximately 1:7 [24] and 1:4 [21]. Extracting the total number of counts attributed to the $\beta\beta$ decay depends greatly on a reliable fit to the background.

The technique utilized in the present Article involves the use of two low-background HPGe detectors operated in a coincidence scheme, the first time such a $\gamma\gamma$ coincidence technique has been applied to the study of $\beta\beta$ decay. In some cases, as in the present one, the reduced detection efficiency of the coincidence measurement can be more than compensated by a corresponding suppression of the associated background. This alternative approach to background reduction relies on the simultaneous detection of the two $\gamma$ rays emitted from the deexcitation of the daughter nuclide. Owing to the unique $\gamma$-ray energies, the coincidence background can be suppressed to such an extent that the signal-to-background ratio may be much higher than in single $\gamma$-ray searches, even in above-ground experiments.
A. TUNL-ITEP Apparatus

Conducted in the Low Background Counting Facility of the Triangle Universities Nuclear Laboratory (TUNL), the experimental work is centered around a $\gamma\gamma$ coincidence apparatus, the cornerstone of which is the operation in coincidence of two high-purity germanium (HPGe) detectors. These large, custom-made HPGe detectors (p-type coaxial) were specially designed to have a large frontal surface area. The size of each HPGe crystal was 8.8 cm in diameter and 5.0 cm in thickness. Each detector was fabricated from low-background materials, and each crystal was coupled to a very-low background cryostat in the J-type configuration, which is used not only to isolate the detector crystal from the preamplifier, the HV filter and LN$_2$ dewar, but also to allow for its easy insertion into an anticoincidence annulus. The two HPGe detectors have 1.8-keV FWHM energy resolution at 1.33 MeV and 0.8-keV resolution at 0.122 MeV, with the relevant peak-to-Compton ratio rated as 80.4.

To suppress the inherent background contributions that result from primary cosmic-ray radiation, secondary cosmogenically-induced radiation and primordial radiation, active and passive shielding techniques were employed. The active shielding consists of anticoincidence counters, which include two plastic plate scintillators (10-cm thickness) and one NaI(Tl) annulus (56-cm length, 35.6-cm diameter with a 12.5-cm hole along the axis of symmetry) into which the source and HPGe detectors are inserted. These counters also veto potential backgrounds arising from unwanted Compton scattering events within the HPGe detectors. The passive shielding includes a lead-brick enclosure and the overhead shielding afforded by the room ceiling and the floors of the building (\(\sim 10\) m w.e.). A schematic view of the apparatus can be seen in Figure 2.

A reliable energy calibration of the HPGe detectors was attained by monitoring known background $\gamma$ peaks (238.6 keV \(^{212}\)Pb), 511.0 keV (annihilation), 609.3 keV \(^{211}\)Bi), and 1460.8 keV \(^{40}\)K). Various performance checks were used to verify the proper operation of the detectors and the electronics over the course of the experiment. For example, daily monitoring of the location of the more prominent background $\gamma$ peaks allowed for any corrections due to gain shifts. Secondly, the sensitivity of the veto system was readily checked by monitoring the strength of known single gamma lines (e.g., 1461-keV $\gamma$ rays from \(^{40}\)K), which in principle are not vetoed, against those emitted in coincidence with additional gamma quanta (e.g., 511-keV annihilation $\gamma$ rays), which will normally be vetoed.

For the study of the $2\nu\beta\beta$ decay of \(^{100}\)Mo to excited states of \(^{100}\)Ru, an enriched metallic disk was fabricated and loaned to TUNL by Russian collaborators from the Institute of Theoretical and Experimental Physics (ITEP). It was enriched in \(^{100}\)Mo to 98.4% and had a mass of 1.05 kg. The disk was approximately 1.1 centimeters thick and 10.6 centimeters in diameter, which

![FIG. 2: (Color online) Schematic view (not to scale) of the TUNL-ITEP $\beta\beta$-decay apparatus. The enriched (98.4%) disk of \(^{100}\)Mo is sandwiched between the two HPGe detectors.](image)

more or less matches the size of the two HPGe detectors. The disk thickness was chosen in order to achieve a suitable balance between maximizing the source mass while limiting efficiency losses due to attenuation and geometry effects. Preceding the study of this enriched disk, a metallic disk of natural molybdenum (1-kg mass, 10-cm diameter, 0.965-cm thickness, 9.6% \(^{100}\)Mo) was studied to better understand potential backgrounds in the coincidence spectra.

B. Efficiency Determination

In order to properly interpret the $\gamma\gamma$ coincidence spectra obtained, a solid understanding of the efficiency of the apparatus to detect $\gamma\gamma$ coincidence events must be achieved. For the purpose of the present study, it was realized that a direct measurement of the efficiency was not only feasible, but it would also provide the most reliable information. Using a source of known strength, the principle behind the efficiency measurement is to observe a deexcitation cascade involving two $\gamma$ rays with energies and angular distributions comparable to those of interest from the $\beta\beta$-decay measurements. As such, the radioactive \(^{102}\)Rh nuclide was judiciously selected for this measurement.

The choice of the \(^{102}\)Rh isotope is quite practical for many reasons. First, the half-life of \(^{102}\)Rh is approxi-
approximately 207 days, which is convenient because the short-lived radioactivities decay away leaving a rather pure sample. Moreover, the source can be used over long periods of time without having to apply significant corrections to a set of measurements performed over a period of a few weeks. Secondly, as shown in Figure 3, following the electron capture of $^{102}$Rh to the $0^+_1$ state of $^{102}$Ru with a branching ratio of 2.68%, the isotope emits two $\gamma$ rays in coincidence with energies (468.6 keV and 475.1 keV) similar to those from the $\beta\beta(0^+ \rightarrow 0^+_1)$ decay of $^{100}$Mo. Since $\beta^+$ decay does not occur, there is no radiation from bremsstrahlung or annihilation, thus making the measurement very simple. At the same time, the probability to reach the $0^+_1$ state via electron capture to higher excited states is small (< 8.8%), hence most of these pairs of $\gamma$ rays (468.6 - 475.1 keV) are emitted without any additional quanta that could interfere with the measurement by a summation effect.

In addition, the angular correlation of the two emitted $\gamma$ rays is identical to those of the $\gamma\gamma$ cascades that follow the $\beta\beta$ decay of $^{100}$Mo to excited $0^+$ final states because it proceeds via the $0^+ \rightarrow 2^+ \rightarrow 0^+$ sequence. This condition arises because the corresponding deexcitation cascade proceeds solely through electric quadrupole (E2) radiation, with no mixing of other multipolarities. Finally, one additional advantage of $^{102}$Rh as the source is the presence of a higher excited $2^+_1$ state at 1103.2 keV of $^{102}$Ru, corresponding to a subsequent $2^+ \rightarrow 2^+ \rightarrow 0^+$ cascade that provides a measure of the detection efficiency for such a deexcitation sequence. Hence, the efficiency for detecting $\beta\beta$ decays to excited $2^+$ states can also be determined, albeit in a more complicated manner because the mixing of multipolarities (M1 and E2) depends on the specific $\gamma$ ray involved (see Figure 4).

A $^{102}$Rh source was produced at TUNL by bombarding a target made from natural ruthenium (31.6% $^{102}$Ru) with a 5-MeV proton beam, thereby generating $^{102}$Rh via (p,n) activation of $^{102}$Ru. Following the production of the $^{102}$Rh source, a small portion of it (0.2 cm × 0.2 cm × 0.1 cm) was isolated for use in the efficiency measurement. Its strength was determined by comparing the intensity of the 475.1-keV gamma line to that of the 661.6-keV gamma line from a calibrated $^{137}$Cs source. To reproduce the photon attenuation in molybdenum that occurs in the $\beta\beta$-decay experiment, the source was placed between thin natural molybdenum disks. Ten disks (10-cm diameter and 0.1-cm thickness) were used to ensure that the overall thickness was the same as the original experiment, thereby creating the same spacing between the two HPGe detectors. The use of these disks also enabled the variation of the position of the $^{102}$Rh source with respect to the molybdenum disks, as a function of cylindrical coordinates (radius, depth, and azimuthal angle) with respect to the center of the detectors.

At each location of the $^{102}$Rh source, the number of $\gamma\gamma$ coincidence events involving the 469 - 475 keV cascade, arising from the decay of $^{102}$Rh to the $0^+_1$ state of $^{102}$Ru, were counted. The results were compared to the total number of decays that actually occurred, as given by the source strength and the branching ratio. Measurements of the $\gamma$ rays emitted from the $^{102}$Rh source were adjusted to corresponding $^{100}$Ru $\gamma$-decay sequences by applying corrections for both the relative efficiency of the HPGe detectors and the photon attenuation in Molybdenum. Figure 4 displays the results for the radial dependence of the efficiency $\varepsilon_{\gamma\gamma}(r)$ to detect the 591 - 540 keV $\gamma\gamma$ coincidence events from the $^{100}$Mo $\beta\beta$ decay. (The notation used henceforth, such as 591 - 540 keV $\gamma\gamma$ coincidence events in which a count in one detector is observed in the energy interval 591 ± 2.5 keV while at the same time a count is registered in the sec-

---

**FIG. 3:** Decay scheme of $^{102}$Rh and the relevant gamma cascades of the excited states of $^{102}$Ru.

**FIG. 4:** (Color online) Calculated angular distribution, $W$, as a function of the angle $\theta_{\gamma_1-\gamma_2}$ between the two $\gamma$ rays emitted in the $\gamma\gamma$ cascade. The solid curve is the angular correlation for all $0^+ \rightarrow 2^+ \rightarrow 0^+$ decay sequences. The dashed (dotted) curve is the correlation between the two $\gamma$ rays emitted from the $2^+_1 \rightarrow 2^+_1 \rightarrow 0^+$ sequence of $^{100}$Ru ($^{102}$Ru).
cascades of interest from excited states in.

FIG. 5: (Color online) The efficiency $\varepsilon_{\gamma\gamma}(r)$ to detect 591 - 540 keV coincidences, measured as a function of the radial distance $r$ from the center of the detectors, and an appropriate fit to the data points.

ond detector in the interval 540 ± 2.5 keV. The notation applies to all possible $\gamma\gamma$ combinations and energies, and the energy interval used (±2.5 keV) is reflective of the detector resolution.

An integration over the $^{100}$Mo source volume yields the total detection efficiency $\varepsilon_{\gamma\gamma}^\text{tot}$, and the results for the $\gamma\gamma$ cascades of interest from excited states in $^{100}$Ru are presented in Table II for the enriched $^{100}$Mo disk. A Monte Carlo simulation was also performed to check the validity of these values. It incorporated all known factors, including the full-energy peak efficiency of the HPGe detectors, the strongly anisotropic angular correlation of the $\gamma$ rays and their attenuation in the sample, and effects of the extended geometry.

III. ANALYSIS

A. Background Considerations

Before proceeding with an analysis of the data from the TUNL-ITEP apparatus, it is important to address potential background contributions that may arise in the $\gamma\gamma$ coincidence spectrum. Although the discussion that follows pertains to the $\beta\beta$ decay of $^{100}$Mo to the $0^+_1$ state of $^{100}$Ru, the principle arguments can be easily extended to decays involving higher excited states (i.e., different $\gamma$-ray energies).

Due to the excellent energy resolution afforded by the HPGe detectors, the primary concern is background processes that can generate an identical gamma decay cascade with no additional quanta. Based on the present criteria to detect excited-state double beta decays by means of the simultaneous detection of the two $\gamma$ rays subsequently emitted via deexcitation, any such background source would in principle be indistinguishable from the desired experimental signature.

Realizing that the cascade involving two, and only two, $\gamma$ rays with energies of 591 keV and 540 keV is unique to the deexcitation of the $0^+_1$ state of $^{100}$Ru, the major background consideration is of any mechanisms other than $\beta\beta$ decay that may populate this excited level. Taking into account the fact that the $\gamma\gamma$ detection efficiency rapidly decreases as one moves away from the center of the $^{100}$Mo disk, as seen in Figure 5, background sources that are of the greatest concern are those originating from within the source itself.

As such, the most relevant background candidate would appear to be a significant flux through the detector setup of cosmic-induced protons, which can in principle result in the $(p,n)$ activation of $^{100}$Mo to bound states in $^{100}$Tc. The subsequent $\beta$ decay of this intermediate nucleus is delayed by the 15.8-s half-life, thereby rendering the anticoincidence counters ineffective in vetoing this process. The branching ratio of the $^{100}$Tc $\beta$ decay to the $0^+_1$ state of $^{100}$Ru is 5.7%. Unfortunately, most of the remainder (93%) of $^{100}$Tc $\beta$ decays proceed to the ground state, which prevents using the $\gamma\gamma$ coincidence spectrum to search for additional gamma decay cascades that might otherwise provide a measure of the $(p,n)$ activation of $^{100}$Mo.

An alternative means of estimating the proton flux in the apparatus and hence estimating the likelihood of background contributions from $^{100}$Mo $(p,n)$ is to search for the very similar $(p,n)$ activation of $^{95}$Mo to the isomer (1/2−, 61-day half-life) of $^{95}$Tc. The electron capture of $^{95m}$Tc populates two levels (1039.2 keV and 786.2 keV) of $^{95}$Mo with large branching ratios (30.1% and 38.0%, respectively). The 1039.2-keV state decays to the ground state 89% of the time via a cascade of two $\gamma$ rays (835.1 keV and 204.1 keV), while the 786.2-keV state proceeds to the ground state with a $\gamma\gamma$ cascade (where $E_{\gamma 1} = 582.1$ keV and $E_{\gamma 2} = 204.1$ keV) 78% of the time. The intermediate 204-keV level has a negligibly short half-life of 0.75 ns. Searching for the 835 - 204 keV and 582 - 204 keV coincidence events provides a convenient method for measuring the proton flux in the laboratory.

To accomplish this study, a metallic disk of natural molybdenum (1-kg mass, 10-cm diameter, 0.965-cm thickness, 15.9% $^{95}$Mo) was investigated in the TUNL-ITEP apparatus for 180 days. Analysis of the data revealed exactly zero 835 - 204 keV coincidence events. For the 582 - 204 keV coincidence, there was no excess of counts observed beyond a fit to the background. Combining these two results allows one to gauge the potential for any contribution of $(p,n)$ activation in the $^{100}$Mo sample, assuming a similar proton flux. The conclusion from the analysis of the $^{95}$Mo study is that $(p,n)$ activation contributes less than one count per year to the important 591 - 540 keV coincidence events of interest.

Another potential background candidate involves sources arising from a significant neutron flux through the detectors and the $^{100}$Mo disk. There exist two known mechanisms that could, in principle, provide unwanted
TABLE I: Calculations of the efficiencies ε^tot (%) to detect γγ cascades resulting from the ββ decay of 100Mo to various excited states of 101Ru.

| Transition | Level (keV) | γ1 − γ2 (keV) | Branching Ratio fb | ε^tot (%) |
|------------|------------|---------------|-------------------|------------|
| 0^- → 0^+_1 | 1130.3 | 590.8 - 539.5 | 1.00 | 0.219 ± 0.022 |
| 0^- → 2^+_2 | 1362.2 | 822.4 - 539.5 | 0.58 | 0.196 ± 0.020 |
| 0^- → 2^+_3 | 1741.0 | 1201.4 - 539.5 | 0.59 | 0.185 ± 0.019 |
| 0^- → 3^+_1 | 2051.5 | 1512.1 - 539.5 | 0.86 | 0.165 ± 0.017 |
| 0^- → 4^+_1 | 2387.4 | 1847.8 - 539.5 | 0.51 | 0.142 ± 0.014 |
|            |           | 1025.2 - 1362.0 | 0.21 | 0.154 ± 0.015 |

γγ coincidence events that would, in practice, be indistinguishable from the ββ decay to excited final states in the present experiment.

First, if a ruthenium impurity resides in or near the 100Mo disk, elastic scattering of neutrons on 100Ru (12.6% abundance) could produce a 591 - 540 keV coincidence that would, in practice, be indistinguishable from the 100Mo ββ decay event to the 0^+_1 state of 100Ru. A measure of the significance of this possible background can be obtained by searching for similar neutron scattering processes, such as 100Mo(n,n'γ) and 76Ge(n,n'γ), where the abundance of 76Ge is 7.44%. The former can result in an excited state of 100Mo that decays to the ground state through a γγ cascade with Eγ1 = 528.2 keV and Eγ2 = 535.6 keV, whereas the latter produces a γγ cascade with Eγ1 = 545.5 keV and Eγ2 = 562.9 keV from the deexcitation of the 76Ge excited state.

During the experimental trial that studied the enriched 100Mo disk, approximately 12 events per year fit the 528 - 536 keV coincidence profile. Also, for the neutron scattering process on the 76Ge nucleus, the 546 - 563 keV coincidence was observed approximately four times per year. Making realistic assumptions about differences in cross section, detection efficiency, and a reasonable estimate on an upper limit of the size of such an impurity within the 100Mo disk, a conservative estimate of the threat of this neutron scattering background amounts to no more than approximately one half of one 591 - 540 keV coincidence event per year.

Secondly, there is a realistic, albeit unlikely, alternative scenario that could result in a coincidence involving 591-keV and 540-keV γ rays that do not originate from the deexcitation of the 0^+_1 state of 101Ru. Rather, following the neutron capture by a 100Mo nucleus, 100Mo(n,γ), the β decay of 101Mo can populate excited states of 101Tc, from which two deexcitation gamma rays (Eγ1 = 540.1 keV and Eγ2 = 590.9 keV) are emitted as part of a four-photon cascade.

In principle, summation and veto effects should rule out the likelihood of any significant contribution from this background candidate. Furthermore, the wide, neutron-broadened peaks that result from the neutron scattering interactions on germanium nuclei are not accompanied by narrow peaks usually ascribed to thermal neutron capture. Thus, the thermal neutron flux is most likely very small. Even so, there is a small, non-zero probability that the two bystander photons could escape undetected, leaving behind a 591-keV photon in one HPGe detector and a 540-keV photon in the other. Conveniently, an analysis of this possibility is readily accomplished as a byproduct of the normal data compilation.

The transition to the 2218-keV level of 101Tc constitutes only 0.4% of 101Mo β decays, and is followed 0.69% of the time by two possible four-photon cascades containing both the 540.1-keV and the 590.9-keV γ rays. A more likely scenario is the β decay to the 1319.6-keV level with a branching ratio of 6.8%, which produces a γγ cascade of two photons with energies of 713.0 keV and 590.9 keV. This γγ cascade is approximately 1000 times more likely to occur as the other four-photon cascades. An analysis of the 455 days of data from the 100Mo run reveals four such 713 - 591 keV coincidence events. Thus, one can place a conservative upper limit of 0.003 counts per year originating from 540 - 591 keV background events from the four-photon deexcitation cascades involved in the β decay of 101Mo. Again, the actual contribution should, however, be considerably less because the two other photons from the four-photon cascades would normally lead to the veto of the signal.

Finally, for reference sake, it was found that truly random coincidences contribute less than 0.0003 counts yr^-1 for the 100Mo experiment (with energies near 600 keV). Moreover, this background should be relatively uniform over the region of interest and would not be specific to the gamma decay cascades relevant to the present study.

B. Decay to the 0^+_1 Excited State

The search for the ββ decay of 100Mo to the 0^+_1 state of 101Ru involves the simultaneous detection of the coincident deexcitation (0^+_1 → 2^+_4 → 0^+_3) γ rays, where Eγ1 = 590.8 keV and Eγ2 = 539.5 keV (see Figure 4). This γγ cascade from the 0^+_1 state of 101Ru has a branching ratio of 100%. The isotopically-enriched (98.4%) 100Mo disk was measured for a period of 455 days in the TUNL-ITSEP apparatus. An inspection of the γ-ray spectra in coincidence with 539.5±2.5 keV and 590.8±2.5...
FIG. 6: (Color online) The $\gamma$-ray spectra in coincidence with 590.8 $\pm$ 2.5 keV (a) and with 539.5 $\pm$ 2.5 keV (b). The 22 observed 591 - 540 keV events (shaded) from the $\beta\beta(0^+ \rightarrow 0^+_1)$ decay of $^{100}$Mo were obtained in 455 days of measuring time.

Deriving an empirical measure of the half-life of the excited-state $\beta\beta$ decay from the observed counting rate is straightforward. The 22 detected events ($\sigma_N = \sqrt{22} = 4.7$) yield $N_{\gamma\gamma} = 19.5 \pm 4.7$ events after subtracting the background, thereby corresponding to a signal-to-background ratio of approximately 8:1. From the sample size (1.05 kg), the counting time (~1.25 yr), and the detection efficiency (0.219%), the calculated decay half-life for the present measurement is

$$T_{1/2}^{(0\nu+2\nu)} = \left[6.0_{-1.1}^{+1.9}(\text{stat}) \pm 0.6(\text{syst})\right] \times 10^{20} \text{ yr.} \quad (1)$$

The asymmetrical statistical uncertainty results from the low number of counts, while the systematic error entails the 10% uncertainty in the detection efficiency. It should be noted that this value for the half-life differs slightly from the previously published value of $5.9_{-1.1}^{+1.7}(\text{stat}) \pm 0.6(\text{syst}) \times 10^{20}$ years for this experiment. The difference arises from a slight modification to the calculated efficiency, the inclusion of some additional data (455 days of counting versus the original 440 days), and a corrected calculation of the statistical error bars.

From the decay half-life presented here, it is possible to extract a value for the nuclear matrix element corresponding to this $2\nu\beta\beta$ transition. Using the phase-space factor $G = 1.64 \times 10^{-19} \text{ yr}^{-1}$ (for the axial-factor coupling constant $g_A = 1.25$), one obtains the nuclear matrix element $M_{2\nu}(0^+_1) = 0.101 \pm 0.013$, as scaled by the electron mass.

The detection scheme is incapable of deciphering the two-neutrino mode from the neutrinoless mode of $\beta\beta$ decay, thus the half-life of Equation 1 is listed as an inclusive ($0\nu + 2\nu$) quantity. Nevertheless, using values for $^{100}$Mo $\beta\beta$ transitions to the ground state of $^{100}$Ru ($T_{1/2}^{2\nu} = 6.75 \times 10^{18} \text{ yr}$ and $T_{1/2}^{0\nu} > 5.5 \times 10^{22} \text{ yr, 90\% C.L.}$), one concludes that the neutrinoless mode constitutes less than 0.012% of $\beta\beta$ transitions to the ground state. In fact, the most recent $^{100}$Mo data from the NEMO3 experiment [27] restrict the neutrinoless $\beta\beta$ decay of $^{100}$Mo to the ground state further: $T_{1/2}^{0\nu} = [7.68 \pm 0.02(\text{stat}) \pm 0.54(\text{syst})] \times 10^{18} \text{ yr and } T_{1/2}^{2\nu} > 3.1 \times 10^{23} \text{ yr (90\% C.L.)}$. Thus, assuming a similar proportion for the excited-state $\beta\beta$ decays, it is reasonable to assume a negligible contribution from $0\nu$ events in the present data. This conclusion is further reinforced by theoretical predictions for the neutrinoless $\beta\beta$ decay of $^{100}$Mo to the $0^+_1$ excited state, with the half-life for this $0\nu\beta\beta(0^+ \rightarrow 0^+_1)$ transition estimated to be: $2.59 \times 10^{26}$ yr [27] and $(0.76 - 1.46) \times 10^{25}$ yr [28] for $\langle m_\nu \rangle = 1 \text{ eV}$.

In Figure 4 the $\gamma$-ray spectra in coincidence with 539.5 $\pm$ 2.5 keV and 590.8 $\pm$ 2.5 keV show a broader range of energies and reveal some interesting features of the background. Both spectra exhibit the presence of some known gamma peaks, such as the annihilation peak (511 keV) and peaks from $^{208}$Tl (583 keV) and $^{214}$Bi (609 keV). As expected, the background at lower energies (< 500 keV) is generally more prominent. This phenomenon is a consequence of two factors: (1) the larger photopeak detection efficiency at lower energies and (2) the contributions at low energies from Compton scattering events. In the latter case, the Compton continuum of a large number of the more prominent primordial background $\gamma$ rays, in addition to those of interest in the present study, lie below 500 keV.

Another interesting feature that was observed in the $\gamma\gamma$ coincidence spectra at higher energies (> 800 keV) is the Compton scattering of at least two known primordial background peaks. The most noticeable instance involved the 1461-keV $\gamma$ ray ($^{40}$K), which is always emitted in single with no accompanying gamma quanta. The Compton scattering of this photon can lead to the deposition of 591 (540) keV in one of the HPGe detectors and the remaining 870 (921) keV in the other HPGe detector, and there are indeed rather prominent peaks at the respective energies of the corresponding spectra of Figure
The same phenomenon occurs for the 1764.5-keV γ-ray ($^{214}$Bi), producing the observed 1225 - 540 keV and 1174 - 591 keV coincidence events. To a lesser extent, there also appear to be some Compton scattering coincidence events corresponding to 1001.0-keV ($^{234m}$Pa) and 1120.3-keV ($^{214}$Bi) γ-rays.

C. Decay to Higher Excited States

In addition to the transition to the excited $0^+_1$ state, the $\beta\beta$ decay of $^{100}$Mo can also populate higher excited states of $^{100}$Ru. In principle, the measurement of these additional transitions is readily accomplished with the TUNL-ITEP apparatus by searching for alternative $\gamma\gamma$ coincidence events associated with the deexcitation of these higher excited states. In the present study, the relevant $^{100}$Ru levels of interest and the corresponding gamma decay cascades are shown in Figure 8.

Since these other processes were not observed in the present experiment, it is useful to briefly mention the procedure for setting a lower limit on a decay half-life. In general, the half-life limit is given by

$$ T_{1/2} > \frac{(\ln 2) N_0 t f_\beta \varepsilon_{\gamma\gamma}^{\text{tot}}}{N_d}, \quad (2) $$

where $f_\beta$ denotes the branching ratio of a given $\gamma\gamma$ cascade, and all relevant values for the detection efficiency $\varepsilon_{\gamma\gamma}^{\text{tot}}$ are taken from Table I. As recommended by the Particle Data Group [29], the factor $N_d$, which represents an upper limit on the number of detected events above background, is determined by a statistical estimator derived for a process that obeys Poisson statistics.

The deexcitation of the higher excited $0^+$ states of $^{100}$Ru at 1741.0, 2051.7, and 2387.2 keV can each occur via two different $\gamma\gamma$ cascades, one involving the transition through the $2^+_1$ state (539.5 keV) and the other through the $2^+_2$ state (1362.0 keV). Hence, the search for $\beta\beta$ decay to these excited states involves monitoring.
the $\gamma$-ray spectrum in coincidence with these two $\gamma$-ray energies. It should be noted that, in addition to these two-photon cascades, there is also a three-photon deexcitation sequence. However, the existing apparatus is not sensitive to cascades involving more than two $\gamma$ rays, thus such decay sequences will not be considered here. At the same time, a search for the $\beta\beta$ decay to the $2_2^+$ excited state is accomplished by searching for coincidence events involving $822.4$-keV and $539.5$-keV $\gamma$ rays, without the emission of any additional gamma quanta.

An analysis of the 455 days worth of data yielded the following results for the $\beta\beta$ decay of $^{100}$Mo to higher excited states. For the transition to the $0^+_2$ state ($1741.0$ keV) of $^{100}$Ru, zero $1201 - 540$ keV coincidence events were detected and one $379 - 1362$ keV event was observed, which corresponds to a counting rate that is consistent with the background. In the case of the transition to the $0^+_2$ state ($2051.7$ keV), a search revealed one $1512 - 540$ keV event (again consistent with the corresponding background rate) and zero $689 - 1362$ keV events. For the transition to the $0^+_4$ state ($2387.2$ keV), zero $1848 - 540$ keV events and zero $1025 - 1362$ keV events were observed. Lastly, a search for $822 - 540$ keV coincidence events, corresponding to the $\beta\beta$ decay to the $2_2^+$ state, yielded zero counts. Taking into consideration the associated $\gamma\gamma$ background rate in the regions of interest, limits on the $\beta\beta$-decay transitions to higher excited states were calculated from Equation 2 and are listed in Table II.

On a more general note, it is interesting to point out that the present $\gamma\gamma$ counting technique represents a powerful tool that could potentially be extended to future, large-scale neutrinoless double beta decay experiments, such as Majorana [35], GENIUS [36], SuperNEMO [37], and CUORE [38], for example. Although $0\nu\beta\beta$ transitions to excited final states have smaller $Q_{\beta\beta}$ values, the emission of fixed-energy, deexcitation $\gamma$ rays opens the possibility of detecting the $0\nu\beta\beta$ decay by way of a multiple, unambiguous coincidence involving the successful detection of both the emitted $\beta$ particles as well as the $\gamma$ rays. Such a multiple coincidence technique would result in a dramatically-reduced background, provided the decay products are detected with high efficiency and with good energy resolution.

Indeed, the information that can be gained from searching for neutrinoless double beta decay to excited final states is just as valuable as that from the transition to the ground state. Furthermore, depending on the particular experimental design and apparatus, it may in principle be possible to incorporate the option to search for $0\nu\beta\beta$ transitions to excited final states, in conjunction with the search for the transition to the ground state, with minimal cost or reconfiguration. Adding this possibility would be considerably advantageous as it would serve to greatly augment the breadth of a given experiment, thereby increasing the potential for discovery.

IV. CONCLUSIONS

The implementation of a $\gamma\gamma$ coincidence counting technique has resulted in a half-life of the $\beta\beta(0^+_1 \rightarrow 0^+_1)$ transition of $^{100}$Mo of $[6.0^{+1.9}_{-1.1}(\text{stat}) \pm 0.6(\text{syst})] \times 10^{20}$ yr, and the corresponding nuclear matrix element is calculated to be $M_{\beta\beta}^2(0^+_1) = 0.101 \pm 0.013$, as scaled by the electron mass. This half-life is in very good agreement with previous results for this transition [21,22], and represents an independent confirmation of this decay. Moreover, the success of the present experiment and the methods used is exemplified by a superior signal-to-background ratio. Although the decay to additional excited final states was not observed in the present measurement, stricter limits on the decay half-lives for these transitions were extracted.

This result is particularly important in the context of the single state dominance hypothesis (SSDH) [31,33]. Using the result for the EC transition $^{100}$Tc $\rightarrow$ $^{100}$Ru [33], a prediction for the $\beta\beta$ decay of $^{100}$Mo to the $0^+_1$ state of $^{100}$Ru has been made: $T_{\beta\beta}^{0\nu} = 4.45 \times 10^{20}$ yr [34]. Unfortunately, this prediction has an accuracy of only 50% due to the uncertainty of the measured EC transition. However, if in the future there is significant improvement in the accuracy of the EC transition, it may be interesting to compare the experimental half-life as presented here to the theoretical prediction to check on the validity of the SSDH.

Acknowledgments

The authors would like to thank Werner Tornow, Christopher Gould, and Frank Avignone, III for their help and advice on the development and execution of this project. This work was supported in part by the U.S. Department of Energy, Office of High Energy and Nuclear Physics, under Grant No. DE-FG02-97ER41033.
TABLE II: A summary of the experimental half-life values, or limits, for the $\beta\beta$ decay of $^{100}$Mo to various excited states of $^{100}$Ru corresponding to a 455-day counting period. Limits are indicated at the 68(90)% C.L. References for previous results are given in square brackets.

| Transition     | Level (keV) | $Q_{\beta\beta}$ (keV) | $T^{(0\nu+2\nu)}_{1/2}$ (×10$^{20}$ yr) | Previous results (×10$^{20}$ yr) |
|----------------|-------------|-------------------------|---------------------------------------|-----------------------------------|
| $0^+ \rightarrow 0^+_1$ | 1130.3      | 1904.0                  | 6.0$^{+1.1}_{-1.1}$ (stat) ± 0.6(syst) | 6.1$^{+1.1}_{-1.1}$ [20]          |
| $0^+ \rightarrow 2^+_2$ | 1362.2      | 1672.1                  | > 54(27)                              | > (13) [20]                       |
| $0^+ \rightarrow 0^+_3$ | 1741.0      | 1293.3                  | > 55(28)                              | > (13) [20]                       |
| $0^+ \rightarrow 0^+_4$ | 2051.7      | 982.6                   | > 42(24)                              |                                   |
| $0^+ \rightarrow 0^+_5$ | 2387.2      | 647.1                   | > 49(25)                              |                                   |

[1] P. C. de Holanda and A. Y. Smirnov, Phys. Rev. D 66, 113005 (2002).
[2] J. N. Bahcall, M. C. Gonzalez-Garcia, and C. Peña-Garay, J. High Energy Phys. 0207, 054 (2002).
[3] M. Maltoni, T. Schwetz, M. A. Tortola, and J. W. F. Valle, Phys. Rev. D 67, 013011 (2003).
[4] KamLAND Collaboration, K. Eguchi, et al., Phys. Rev. Lett. 90, 021802 (2003).
[5] SNO Collaboration, S. N. Ahmed, et al., Phys. Rev. Lett. 92, 181301 (2004).
[6] S. M. Bilenky, hep-ph/0403245.
[7] S. Pascoli, S. T. Petcov, and W. Rodejohann, Phys. Lett. B 558, 141 (2003).
[8] S. Pascoli and S. T. Petcov, Phys. Lett. B 580, 280 (2004).
[9] S. R. Elliott and P. Vogel, Annu. Rev. Nucl. Part. Sci. 52, 115 (2002).
[10] S. R. Elliott and J. Engel, J. Phys. G 30, R183 (2004).
[11] A. S. Barabash, Phys. At. Nucl. 67, 438 (2004).
[12] F. T. Avignone III, Nucl. Phys. B (Proc. Suppl.) 143, 233 (2005).
[13] J. Suhonen, Phys. Lett. B 607, 87 (2005).
[14] V. A. Rodin, A. Faessler, F. Simkovic, and P. Vogel, nucl-th/0503063.
[15] A. S. Barabash, Czech. J. Phys. 52, 567 (2002).
[16] A. Griffiths and P. Vogel, Phys. Rev. C 46, 181 (1992).
[17] M. Aunola and J. Suhonen, Nucl. Phys. A 602, 133 (1996).
[18] J. Suhonen and O. Civitarese, Phys. Rep. 300, 123 (1998).
[19] A. S. Barabash, JETP Lett. 51, 207 (1990).
[20] A. S. Barabash et al., Phys. Lett. B 345, 408 (1995).
[21] A. S. Barabash, R. Gujrirarán, F. Hubert, P. Hubert, and V. I. Umatov, Phys. At. Nucl. 62, 2039 (1999).
[22] A. S. Barabash, F. Hubert, P. Hubert, and V. I. Umatov, JETP Lett. 79, 10 (2004).
[23] L. De Braeckeleeer, M. Hornish, A. Barabash, and V. Umatov, Phys. Rev. Lett. 86, 3510 (2001).
[24] A. De Silva, M. K. Moe, M. A. Nelson, and M. A. Vient, Phys. Rev. C 56, 2451 (1997).
[25] H. Ejiri et al., Phys. Rev. C 63, 065501 (2001).
[26] NEMO Collaboration, R. Arnold, et al., JETP Lett. 80, 377 (2004).
[27] J. Suhonen, Nucl. Phys. A 700, 649 (2002).
[28] F. Simkovic, M. Nowak, W. A. Kamiński, A. A. Raduta, and A. Faessler, Phys. Rev. C 64, 035501 (2001).
[29] S. Eidelman et al., Phys. Lett. B 592, 1 (2004), URL http://pdg.lbl.gov.
[30] J. Abad, A. Morales, R. Nuñez-Lagos, and A. F. Pacheco, Ann. Fis. A 80, 9 (1984).
[31] O. Civitarese and J. Suhonen, Phys. Rev. C 58, 1535 (1998).
[32] O. Civitarese and J. Suhonen, Nucl. Phys. A 653, 321 (1999).
[33] A. Garcia et al., Phys. Rev. C 47, 2910 (1993).
[34] F. Simkovic, P. Domin, and S. V. Semenov, J. Phys. G 27, 2233 (2001).
[35] C. E. Aalseth et al., hep-ex/0201021.
[36] H. V. Klapdor-Kleingrothaus et al., hep-ph/0103074.
[37] A. S. Barabash and NEMO Collaboration, Czech. J. Phys. 52, 575 (2002).
[38] C. Arnaboldi et al., hep-ex/0212053.