Double-electron capture on ^{112}Sn to the excited 1871 keV state in ^{112}Cd: A possible alternative to double-β decay

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We report the first use of a coincidence technique to study neutrinoless double-electron capture (0νECEC) to an excited state in the daughter nucleus. We investigated 0νECEC by ^{112}Sn leading to the possibly degenerate 1871 keV excited state in ^{112}Cd by searching for its deexcitation γ rays of 1253 keV and 618 keV in coincidence. The experiment was performed at ground level. A sample of 3.91 g of tin enriched to 99.5% in ^{112}Sn was placed between two high-purity germanium γ-ray detectors. In order to enhance the sample material, rods of natural tin totaling 1.2 kg (natural abundance 0.97% ^{112}Sn) surrounded the cylindrical surface of our two γ-ray detectors. After an exposure of 1.59 kg $\times$ days of ^{112}Sn, no decays were observed. From this null result we determine a lower limit for the half-life time of $T_{1/2} > 2.7(1.3) \times 10^{19}$ yr (68%(90%) CL). We hope to achieve a sensitivity in the 10^{23} to 10^{24} yr range with a sample of a few kg of ^{112}Sn and improved γ-ray detectors in an underground facility.

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I. INTRODUCTION

In 1955 Winter [1] pointed out the unique role of neutrinoless double-electron capture (0νECEC) to very special daughter nuclei which have excited states degenerate with the parent nucleus. He wrote: "[T]he existence of a state to which the transition can go with only double capture without real neutrino emission would be a monumental coincidence." In 1981 Georgi, Glashow, and Nussinov [2] rejected the importance of 0νECEC reactions, writing: "We know of no instance where this process can realistically be detected." However, only two years later Bernabeu, De Rujula, and Jarlskog [3] discussed 0νECEC reactions as a tool to measure the electron neutrino mass. They wrote: "In some cases where the daughter nucleus is excited, the neutrinoless decay may be enhanced by its proximity to a virtual resonance. We identify the ^{112}Sn $\rightarrow$ ^{112}Cd transition as a good candidate." They found that the sensitivity of this reaction to the neutrino mass is comparable to that of neutrinoless double-beta ($0\nu\beta\beta$) decay. More recently, Sujkowski and Wycech [4] studied the resonant enhancement for a specific 0νECEC reaction (^{152}Gd $\rightarrow$ ^{152}Sm), but, in general terms, also referred to the possibility of transitions to excited final states in daughter nuclei. Very recently, Frekers [5] proposed to search for the 1204 keV resonance transition in the case of the 0νECEC reaction ^{74}Se $\rightarrow$ ^{74}Ge, where the 1204 keV excited state in ^{74}Ge is degenerate with the ^{74}Se ground state within the uncertainty in the atomic mass difference of 2.3 keV (assuming ECEC of L-shell electrons). Subsequently, Barabash *et al.* [6] carried out such a search. They found a lower limit of $T_{1/2} > 5.5 \times 10^{18}$ yr (90% CL) for the 0νECEC transition to this second excited 2^+ state in ^{74}Ge. Also in 2007, Dawson *et al.* [7] reported a lower limit of $T_{1/2} > 1.6 \times 10^{18}$ yr (90% CL) for the 0νECEC reaction on ^{112}Sn in their search for the decay of the 1871 keV third excited 0^+ state in ^{112}Cd, which is degenerate with the ^{112}Sn ground state within the uncertainty in the atomic mass difference of 5 keV (assuming ECEC of K-shell electrons).

Assuming the decay of the 1204 keV state in ^{74}Ge or the decay of the 1871 keV state in ^{112}Cd is observed and it can be shown that these states are not produced by cosmic-ray induced reactions, a clear signature for the 0νECEC reaction would be obtained. As a result, the Majorana nature of the electron neutrino would be established, and a value for the effective neutrino mass could be obtained, provided the resonance enhancement can be reliably calculated. These are the compelling arguments why the study of 0νECEC reactions is conceivably a viable alternative to $0\nu\beta\beta$ studies.

Here we describe our search for the 0νECEC reaction on ^{112}Sn using the TUNL-ITEP double-beta decay apparatus [8]. This work was underway when we learned about [7]. A short report about our work was presented in [9]. When our paper was ready for submission a final literature search revealed the paper of Barabash *et al.* [10] on tin isotopes.

II. THE TUNL-ITEP DOUBLE-BETA DECAY APPARATUS

The TUNL-ITEP double-beta decay apparatus [8] was originally designed to measure $2\nu\beta\beta$ half-life times for transitions to excited final states in daughter nuclei. It was successfully operated at ground level to measure $T_{1/2}$ of ^{100}Mo to the first excited 0^+ state in ^{100}Ru by detecting the two deexcitation γ rays of 591 keV and 540 keV in coincidence. As seen in Fig. 1, a 1.05 kg disk (10.5 cm in diameter and 1.0 cm thick) of molybdenum enriched to 98.3% in ^{100}Mo was sandwiched between two high-purity germanium (HPGe) detectors (8.8 cm in diameter and 5.0 cm long). These detectors were surrounded by a NaI annulus (54 cm long, 12.5 cm inner diameter, and 10 cm wall thickness) veto counter which in turn was surrounded by a massive enclosure built of lead bricks. Due to the combination of the coincidence requirement and active shielding, half-life times up to 10^{22} yr can be measured

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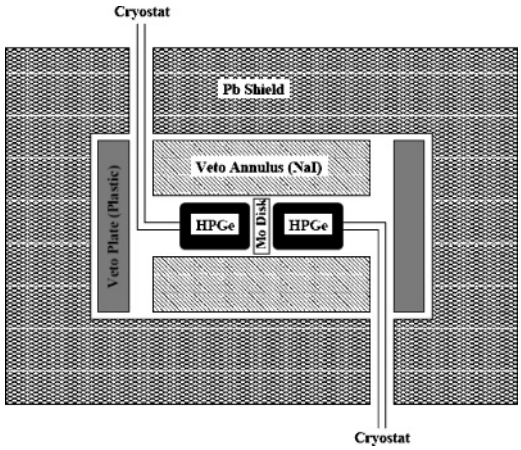

FIG. 1. Setup diagram of the TUNL-ITEP double-beta decay apparatus. See text for details.

with the TUNL-ITEP double-beta decay apparatus. Details are given in [8].

III. SEARCH FOR 0νECEC WITH AN ISOTOPICALLY ENRICHED ^{112}Sn SAMPLE

Figure 2 summarizes the parts of the level scheme of ^{112}Cd which are of interest to the present work. We searched for the decay of the 0_3^+ state located at 1871 keV by detecting 1253 keV and 618 keV γ rays in coincidence. We report our result obtained after 102 days of measurement. We placed 3.91 g of tin enriched to $(99.5 \pm 0.2)\%$ in ^{112}Sn between the two HPGe detectors. Considering the small natural abundance of ^{112}Sn (0.97%), this corresponds to 0.40 kg of natural tin, about one third of the amount used in [7]. Our ^{112}Sn target consisted of two strips 12.2 cm and 7.5 cm long, both with 0.05 cm thickness and 1.6 cm width. These strips were folded once and taped crosswise to the front face at the center of one of our HPGe detectors.

Because the separation of the two γ-ray detectors was only 0.2 cm compared to the 1.1 cm used in [8] and [11], we measured the coincidence efficiency for this new geometry following the procedure described in [11]. Figure 3 shows the measured coincidence efficiency as a function of radius

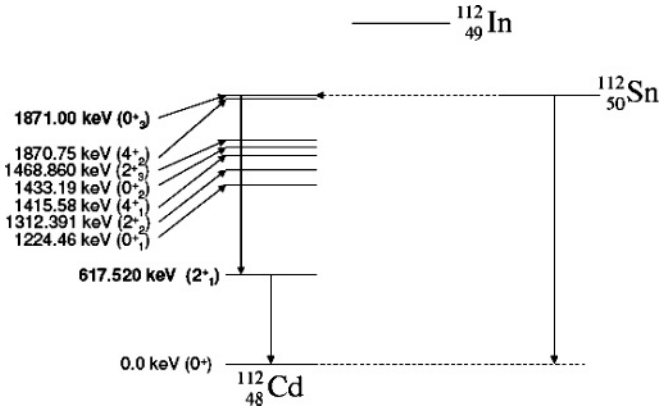

FIG. 2. Level scheme of ^{112}Cd, the daughter nucleus of the ECEC process of ^{112}Sn.

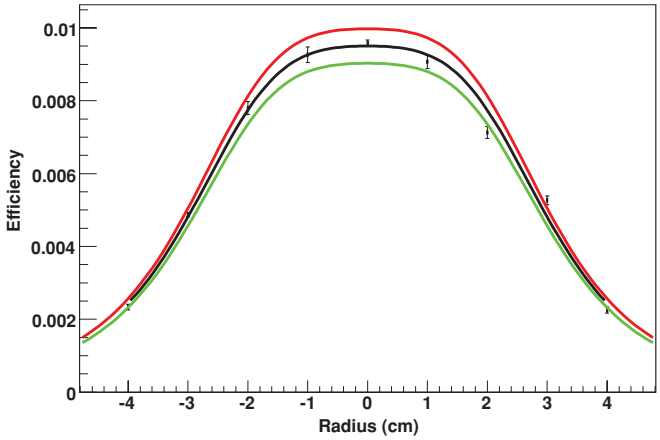

FIG. 3. (Color online) Radial dependence of the coincidence efficiency of the present two HPGe detector setup. The solid curves are a fit curve (solid black) and curves representing the systematic errors of $\pm 5\%$.

measured from the center on the front face of our γ-ray detectors for 618 keV/1253 keV γ-ray pairs. Because the detection efficiency of HPGe detectors is energy dependent, the data obtained with a ^{102}Rh source (469 keV/475 keV, $0^+ \rightarrow 2^+ \rightarrow 0^+ \gamma$-ray pair) were corrected for the difference in relative detection efficiency. The relative efficiencies of our detectors were measured with a ^{152}Eu source while the absolute efficiencies were determined with a ^{137}Cs source of known source strength ($\pm 3\%$). The total coincidence efficiency for the γ-ray pair of interest using our ^{112}Sn target was found to be $(0.85 \pm 0.04)\%$.

In order to increase the amount of target material, we surrounded the cylindrical surface of our two HPGe detectors by 55 rods of natural tin (purity 99.999%), each 10 cm long and 0.6 cm in diameter, resulting in a total mass of 15.7 g of ^{112}Sn. Although this additional amount of ^{112}Sn compares favorably to the 3.91 g of enriched material, one has to take the coincidence efficiency for this special geometry into account. For this purpose, we again followed the approach described in [11] and measured the coincidence efficiency along the length of the rods surrounding the two detectors using our ^{102}Rh source. After the appropriate corrections for attenuation and difference in γ-ray energies, a total coincidence efficiency of $\epsilon_{\gamma\gamma} = (0.22 \pm 0.01)\%$ was obtained. This smaller efficiency results in only a 5% increase in effective ^{112}Sn target mass compared to just the enriched sample.

Figure 4 shows a "singles" spectrum obtained with our HPGe Det. 1 not in coincidence with Det. 2. The measurement was made on ground level for 102 d. Here, the NaI veto surrounding the two detectors was turned on to reject background events, including cosmic-ray induced events in the γ-ray detectors. The strong line seen at 392 keV is due to the decay of ^{113}Sn ($T_{1/2} = 115$ d) which is a contaminant in our sample, resulting from prior exposure to neutron beams. Due to our 11 bit ADC, the spectrum is limited to γ-ray energies up to 2.0 MeV. This spectrum exhibits all the lines which are typical for standard HPGe detectors. In order to provide more details, Fig. 5 presents the same spectrum as Fig. 4, with the regions of interest surrounding 618 keV, 1253 keV, and 1888.5 keV. There

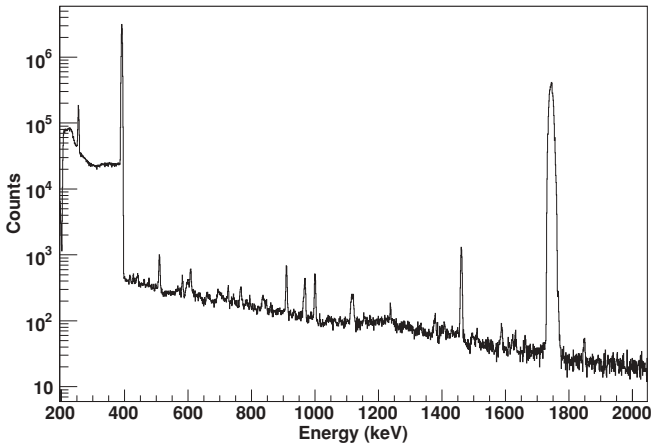

FIG. 4. Energy spectrum ("singles") of ^{112}Sn sample obtained with Det. 1. The strong γ-ray line at 392 keV is due to a ^{113}Sn contamination of the 99.5% enriched ^{112}Sn sample. The large peak near 1750 keV is from the pulser. The threshold is set at 200 keV.

are no peaks visible at these particular γ-ray energies. We will return to the singles spectra obtained in the present work in Sec. IV. Instead, here we will first focus on the main objective of our experimental studies, the search for the simultaneous detection of the 618 keV/1253 keV γ-ray pair.

Figure 6 gives a two-dimensional representation of energy deposited in Det. 1 versus energy deposited in Det. 2. The strong diagonal lines with negative slope are due to 1461 keV, 1765 keV, and 2615 keV background γ rays sharing their energy between the two HPGe detectors. The events in the upper right corner are due to a pulser used for monitoring purposes and dead-time determination. Zooming in onto the regions of interest, i.e., 618 keV in Det. 1 and 1253 keV in Det. 2 and 1253 keV in Det. 1 with 618 keV in Det. 2, we obtained the two energy distributions shown in Fig. 7. There are no events at the energy combinations of interest. These spectra were used to determine a lower limit for the half-life time of ^{112}Sn for the 0νECEC to the excited 1871 keV state in ^{112}Cd using our measured coincidence efficiency. From the 618 keV/1253 keV data we obtained the value of $T_{1/2} > 2.7(1.3) \times 10^{19}$ yr (68(90)%CL). This result is based on 1.59 kg $\times$ days of exposure and represents an improvement by a factor of eight over the value reported in [7]. There, 1.24 kg of natural tin was used and the total exposure was 43.3 kg $\times$ days, which corresponds to 0.42 kg $\times$ days of measurement with a pure ^{112}Sn sample. In contrast to [7], our result obtained with the coincidence technique is almost background free. Background will become significant only once we increase our measuring time or sample material by at least two orders of magnitude. Unfortunately, a larger isotopically enriched ^{112}Sn sample was not available for our studies as the sample used in the present work is to our knowledge the largest isotopically enriched ^{112}Sn sample that currently exists.

IV. ANALYZING SINGLES ENERGY SPECTRA

Although it is not our long-term goal to determine $T_{1/2}$ for 0νECEC by ^{112}Sn to the excited 1871 keV 0^+ state in ^{112}Cd

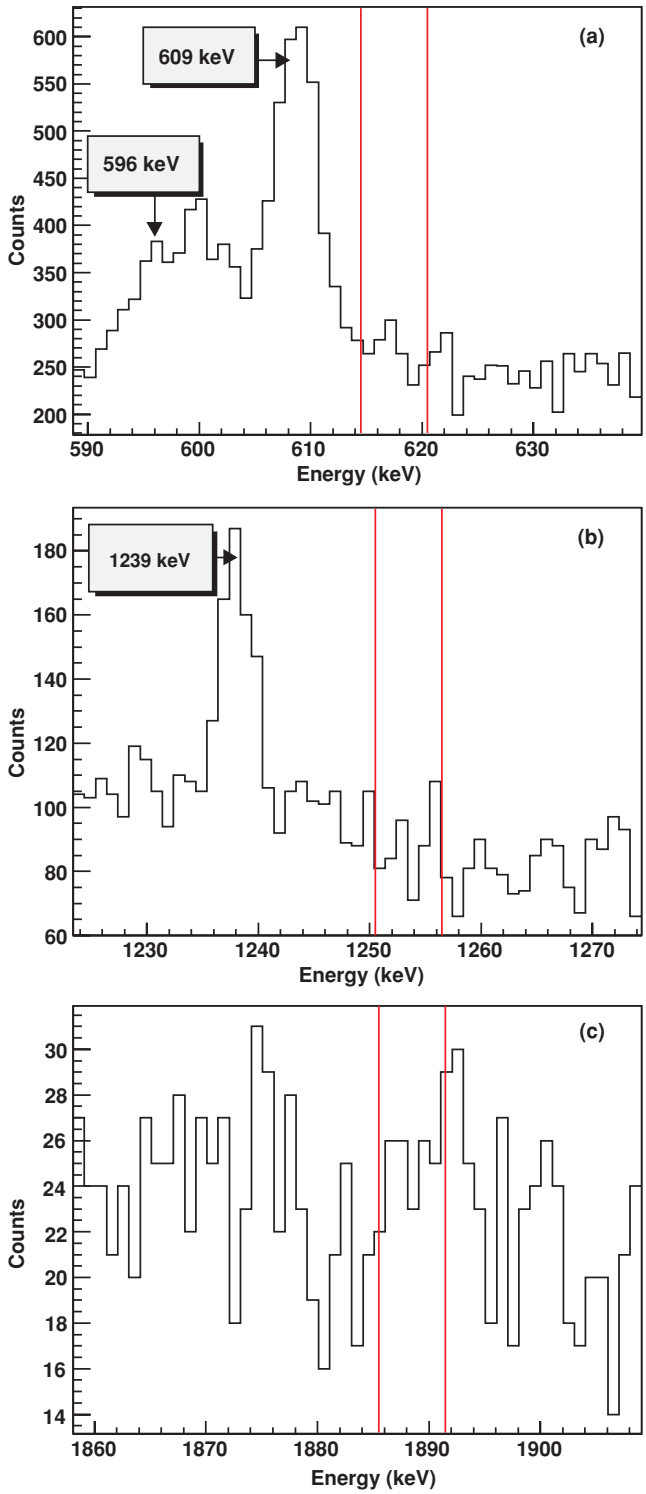

FIG. 5. (Color online) Zoomed in view of Fig. 4. Each section shows an expanded view of the energy regions of interest around 618 keV, 1253 keV, and 1889 keV.

by analyzing singles spectra, at present it is meaningful to analyze these spectra as well. Due to the large efficiency of our individual HPGe detectors compared to their coincidence efficiency, even a poor peak-to-background ratio can provide useful results. Therefore, in this section we concentrate on

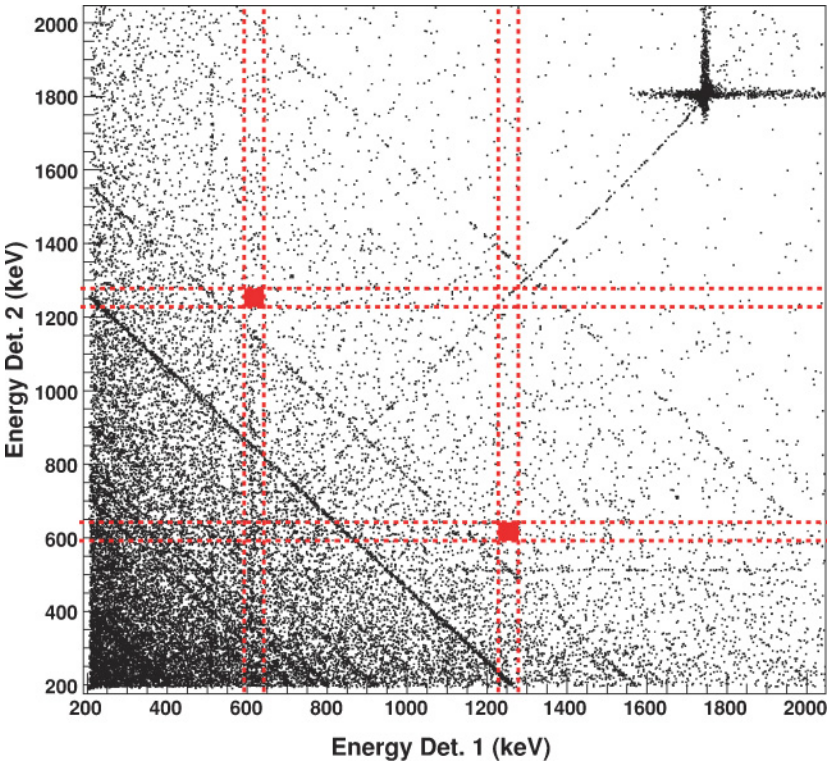

FIG. 6. (Color online) Two-dimensional spectrum of Det. 1 versus Det. 2. The two squares indicate the two energy regions of interest. See text for details.

the analysis of the "singles" spectra obtained with Det. 1 and Det. 2.

We first consider 0νECEC by ^{112}Sn to the ground state of ^{112}Cd, which proceeds via the emission of an internal monoenergetic bremsstrahlung photon of 1888.5 keV (Q value minus K- and L-shell binding energies of ^{112}Cd). Inspecting the spectrum shown in Fig. 5(c) for Det. 1 and the equivalent spectra for Det. 2 we observed no indication of a peak in the energy region of interest. From this null result, the lower limit of $T_{1/2} > 3.4(2.1) \times 10^{18}$ yr (68(90)% CL) has been deduced for this transition. This limit is a factor of two higher than that reported in [7] of 0.99×10^{18} yr (90% CL).

The decay of ^{112}Sn to excited states in ^{112}Cd usually includes the 2_1^+ state at 618 keV. As a result, a search for this γ-ray line places a half-life limit on all excited state transitions, provided the relevant branching ratios are taken into account. On the other hand, because the peak-to-background ratio is energy dependent, a search for the 1253 keV transition might provide a more stringent limit on the 618 keV transition than the direct search focusing on this specific energy. This is especially true in our case where the 618 keV γ-ray line is so close to the ^{73}Ge(n, γ)(596 keV) and ^{214}Bi (609 keV) lines, as can be seen in Fig. 5(a). Therefore, we limit our search to the γ-ray line at 1253 keV. A representative singles spectrum

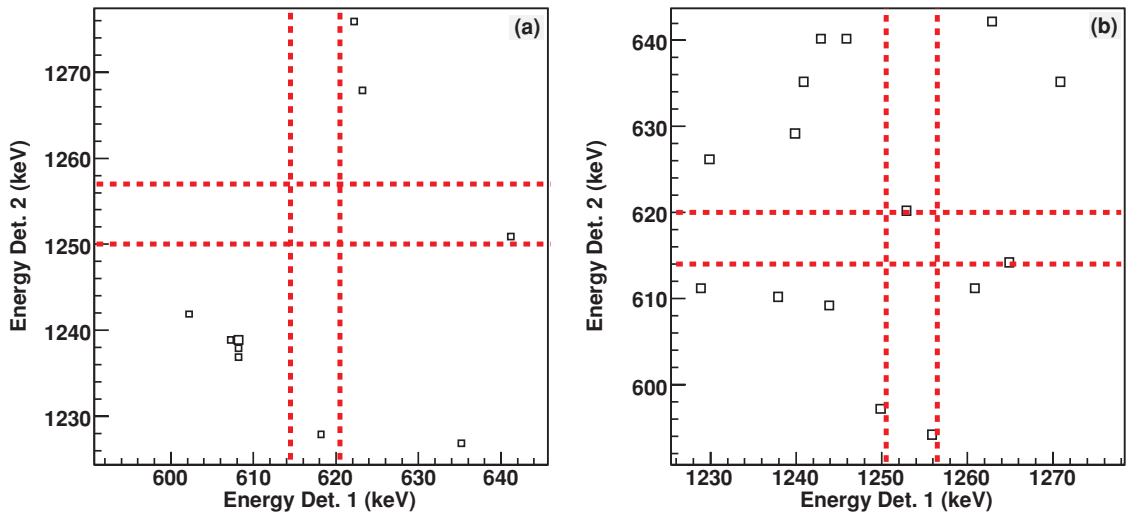

FIG. 7. (Color online) (a) Expanded view of energy region centered around 618 keV in Det. 1 and 1253 keV in Det. 2. (b) same as (a) but centered around 1253 keV in Det. 1 and 618 keV in Det. 2.

obtained with Det. 1 is shown in Fig. 5(b). No excess of counts is observed in the region of interest.

From the two individual singles spectra we determined a lower limit for the 0νECEC reaction on ^{112}Sn to the excited 1871 keV state in ^{112}Cd of $T_{1/2} > 2.0(1.2) \times 10^{18}$ yr (68%(90)% CL). This limit, based on the 1253 keV transition, is comparable to that of 1.4×10^{18} yr (90% CL) obtained in [7], but is a factor of about ten worse than our limit obtained with the coincidence technique.

V. SUMMARY AND CONCLUSION

For the first time the coincidence technique of [8] has been used in ECEC studies. We employed this powerful technique in searches for the 0νECEC reaction on ^{112}Sn to the excited and possibly degenerate 0^+ state of ^{112}Cd at 1871 keV by searching for 1253 keV/618 keV deexcitation γ-ray pairs. No such transitions have been observed in our low background experimental approach. Also for the first time, an enriched tin sample (99.5%^{112}Sn) was used for 0νECEC searches. Our null result corresponds to a half-life time of $T_{1/2} > 1.3 \times 10^{19}$ yr (90% CL). This result is a factor of eight larger than the limit reported in [7], but a factor of about seven worse compared to the very recent result of Barabash *et al.* [10]. We also analyzed our singles spectra focusing on the 1889 keV bremsstrahlung photon emission and on the 1253 keV γ-ray transition. Again, no enhancement above background was observed in the energy regions of interest. From these null results a lower limit of $T_{1/2} > 2.1 \times 10^{18}$ yr was obtained for the internal monoenergetic bremsstrahlung photon emission, and a lower limit of $T_{1/2} > 1.2 \times 10^{18}$ yr for the 0νECEC to the 1871 keV state in ^{112}Cd. Either result is comparable within a factor of two to those reported in [7].

The result of Barabash *et al.* [10] was obtained from the analysis of a singles spectrum using a natural tin sample with an equivalent exposure of 3.6 kg $\times$ days of ^{112}Sn compared to our 1.6 kg $\times$ days. Although their half-life time limit is a factor of seven better than that obtained in our study, there is a substantial amount of background associated with the singles spectrum, which will hinder the usefulness of the singles approach if one tries to improve the limit by orders of magnitude. In contrast, the coincidence approach is far from background-limited and has the potential of providing a much higher sensitivity than can be obtained from a singles spectrum in the long run. Furthermore, the detection of γ rays at 1253 keV or 618 keV in a singles spectrum is no guarantee that the 0νECEC process of ^{112}Sn has in fact been observed. Due to the far–reaching consequences associated with the possible detection of the 0νECEC it is necessary to detect these two γ-ray transitions in coincidence. These are the two main differences between the standard singles spectrum approach of Barabash *et al.* and the coincidence approach reported here.

Of course, the importance of the ^{112}Sn 0νECEC reaction as a means of determining fundamental neutrino properties rests solely on the degree of degeneracy between the 0^+ ground state of ^{112}Sn and the 3rd excited 0^+ state of ^{112}Cd at 1871.00 keV [12] (assuming K-shell capture). Presently, the mass difference between ^{112}Sn and ^{112}Cd is known to 5 keV [13], while the energy of the 1871.00 keV state is known to 190 eV [12] or 130 eV [14]. Therefore, it is crucial to determine the mass difference between ^{112}Sn and ^{112}Cd more accurately before a large scale search for the 0νECEC on ^{112}Sn is pursued. Very recently, Rahaman *et al.* [15] using the JYFLTRAP [16] have measured the double-beta decay Q-values of ^{100}Mo and ^{76}Ge to an accuracy of better than 200 eV. The same group has also measured the Q-values of the ^{112}Sn-^{112}Cd pair to the same accuracy, but a final result has not been released yet [17]. According to Bernabeu *et al.* [3], a degeneracy of 300 eV would increase the 0νECEC rate of ^{112}Sn by a factor of about 250 compared to a degeneracy of 5 keV. If it turns out that the degeneracy is in the 300 eV energy range then it is extremely important to improve both the mass difference measurement and the determination of the excitation energy of the 1871.00 keV state in ^{112}Cd. Recently, Redshaw *et al.* [18] reported on a measurement of the mass and double-beta decay Q-value of ^{136}Xe with an accuracy in the mass measurement of 10 eV. According to Ref. [19], with special effort the Florida State Penning trap group can measure the mass difference of stable nuclei to an accuracy of about 3 eV. Clearly, such an accuracy is out of reach for the measurement of the excitation energy of the 1871.00 keV state. We estimate that in a dedicated experiment the excitation energy of the 1871.00 keV state can be measured to an accuracy of about 40 eV. Assuming a total uncertainty of the degree of degeneracy of 50 eV the calculations of Bernabeu *et al.* imply an enhancement factor of about four orders of magnitude compared to a degeneracy of 5 keV, making the 0νECEC rate about a factor of three larger than the ordinary 2νECEC rate of ^{112}Sn [3]. It is the potential enhancement factor of about 10^4 that makes the 0νECEC search on ^{112}Sn to the 0^+ excited state at 1871.00 keV so fascinating.

We are currently in the process of recommissioning our apparatus after it was moved to the Kimballton mine in Virginia. There, with 1450 meters water equivalent overburden, we are continuing our present work on ^{112}Sn. To increase our total mass of ^{112}Sn we have sandwiched a disk of natural tin with 10.7 cm diameter and 1.0 cm thickness in addition to our 3.91 g of highly enriched ^{112}Sn. Although this disk will attenuate any γ-ray pairs emitted by the enriched sample and, in addition, will reduce our coincidence efficiency due to the increased separation of our two HPGe detectors, the net effect will still provide an increase in effective ^{112}Sn mass by 15% over that of the enriched sample alone. Furthermore, we are exploring ways of acquiring more sample material enriched in ^{112}Sn.

We are confident that the coincidence approach used in the present work is capable of reaching sensitivity limits in the range of 10^{23} to 10^{24} yr for 0νECEC searches to the excited 1871 keV state in ^{112}Cd, provided a 2 kg disk of highly enriched ^{112}Sn will become available and more efficient and radiopure HPGe detectors will be employed. A further increase in sensitivity will require a large plate of ^{112}Sn observed by an array of HPGe detectors in a deep underground facility. With such improvements 0νECEC searches to degenerate excited states in daughter nuclei could become competitive to $0\nu\beta\beta$ decay searches.

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