Search for β^+ EC and ECEC processes in ¹¹²Sn

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Limits on $\beta^+\text{EC}$ (here EC denotes electron capture) and ECEC processes in ¹¹²Sn have been obtained using a 380 cm³ HPGe detector and an external source consisting of 53.355 g enriched tin (94.32% of ¹¹²Sn). A limit with 90% C.L. on the ¹¹²Sn half-life of 4.7×10^{20} y for the ECEC(0v) transition to the 0^+_3 excited state in ¹¹²Cd (1871.0 keV) has been established. This transition is discussed in the context of a possible enhancement of the decay rate by several orders of magnitude given that the ECEC(0v) process is nearly degenerate with an excited state in the daughter nuclide. Prospects for investigating such a process in future experiments are discussed. The limits on other β^+ EC and ECEC processes in ¹¹²Sn were obtained on the level of (0.6–8.7) × 10²⁰ y at the 90% C.L.

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I. INTRODUCTION

Interest in neutrinoless $\beta\beta$ decay has seen a significant renewal in recent years after evidence for neutrino oscillations was obtained from the results of atmospheric, solar, reactor, and accelerator neutrino experiments (see, for example, the discussions in Refs. [1-3]). These results are impressive proof that neutrinos have a nonzero mass. However, the experiments studying neutrino oscillations are not sensitive to the nature of the neutrino mass (Dirac or Majorana) and provide no information on the absolute scale of the neutrino masses, because such experiments are sensitive only to the difference of the masses, Δm^2 . The detection and study of $0\nu\beta\beta$ decay may clarify the following problems of neutrino physics (see discussions in Refs. [4–6]): (i) neutrino nature, whether the neutrino is a Dirac or a Majorana particle, (ii) absolute neutrino mass scale (a measurement or a limit on m_1), (iii) the type of neutrino mass hierarchy (normal, inverted, or quasidegenerate), and (iv) CP violation in the lepton sector (measurement of the Majorana CP-violating phases). At the present time only limits on the level of $\sim 10^{24}$ – 10^{25} yr for half-lives and \sim 0.3–1 eV for effective Majorana neutrino mass $\langle m_{\nu} \rangle$ have been obtained in the best modern experiments (see recent reviews, Refs. [7–9]).

The $\beta\beta$ decay can proceed through transitions to the ground state as well as to various excited states of the daughter nucleus. Studies of the latter transitions provide supplementary information about $\beta\beta$ decay.

Most $\beta\beta$ -decay investigations have concentrated on the $\beta^{-}\beta^{-}$ decay. Much less attention has been given to the investigation of $\beta^{+}\beta^{+}, \beta^{+}EC$, and ECEC processes (here EC denotes electron capture). There are 34 candidates for these processes. Only 6 nuclei can undergo all of the abovementioned processes, 16 nuclei can undergo $\beta^{+}EC$ and ECEC, and 12 nuclei can undergo only ECEC. Detection of the neutrinoless mode in the above processes enables one to determine the effective Majorana neutrino mass $\langle m_{\nu} \rangle$ and parameters of right-handed current admixture in electroweak interaction ($\langle \lambda \rangle$ and $\langle \eta \rangle$). Detection of the two-neutrino mode in the above processes lets one determine the magnitude of the nuclear matrix elements involved, which is very important in view of the theoretical calculations for both the 2ν and the 0ν modes of $\beta\beta$ decay. Interestingly, it was demonstrated in Ref. [10] that if the $\beta^{-}\beta^{-}(0\nu)$ decay is detected, then the experimental limits on the $\beta^{+}EC(0\nu)$ half-lives can be used to obtain information about the relative importance of the Majorana neutrino mass and right-handed current admixtures in electroweak interactions.

The $\beta^+\beta^+$ and β^+EC processes are less favorable because of smaller kinetic energy available for the emitted particles and Coulomb barrier for the positrons. However, an attractive feature of these processes from the experimental point of view is the possibility of detecting either the coincidence signals from four (two) annihilation γ rays and two (one) positrons or the annihilation γ rays only. It is difficult to investigate the ECEC process because one detects only the low-energy x rays. It is also interesting to search for transitions to the excited states of daughter nuclei, which are easier to detect given the cascade of higher energy γ 's [11]. In Ref. [12] it was first mentioned that in the case of $ECEC(0\nu)$ transition a resonance condition can exist for transition to the "right energy" of the excited level for the daughter nucleus; here the decay energy is close to zero. In 1982 the same idea was proposed for the transition to the ground state [13]. In 1983 this possibility was discussed for the transition of 112 Sn to 112 Cd (0⁺; 1871 keV) [14]. In 2004 the idea was reanalyzed in Ref. [15] and new resonance conditions for the decay were formulated. The possible enhancement of the transition rate was estimated as $\sim 10^6$ [14,15]. This means that this process starts to be competitive with $0\nu\beta\beta$ decay for the neutrino mass sensitivity and is interesting to check experimentally. There are several candidates for which resonance transition, to the ground (152 Gd, 164 Eu, and 180 W) and to the excited states (74 Se, 78 Kr, 96 Ru, 106 Cd, 112 Sn, ¹³⁰Ba, ¹³⁶Ce, and ¹⁶²Er) of daughter nuclei, exists [15,16]. The precision needed to realize this resonance condition is well below 1 keV. To select the best candidate from the above list one must know the atomic mass difference with an accuracy

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better than 1 keV and such measurements are planned for the future. Recently the experimental search for such a resonance transition in ⁷⁴Se to ⁷⁴Ge (2⁺; 1206.9 keV) was performed yielding a limit of $T_{1/2} > 5.5 \times 10^{18}$ yr [17]. Very recently ¹¹²Sn was investigated [18–20]. The more strong limit of $T_{1/2} > 0.92 \times 10^{20}$ yr was obtained for the transition to the 0⁺ state at 1871 keV with the 4-kg natural tin sample [18].

In this article the results of an experimental investigation of the β^+ EC and ECEC processes in ¹¹²Sn using the enriched tin sample are presented.

II. EXPERIMENTAL

The experiment was performed in the Modane Underground Laboratory at a depth of 4800 m w.e. The enriched tin sample was measured using a 380 cm³ low-background HPGe detector.

The HPGe spectrometer is a p-type crystal with the cryostat, endcap, and majority of mechanical components made of a very pure Al-Si alloy. The cryostat has a J-type geometry to shield the crystal from radioactive impurities in the dewar. The passive shielding consisted of 4 cm of Roman-era lead and 10 cm of OFHC copper inside 15 cm of ordinary lead. To remove ²²²Rn gas, one of the main sources of the background, a special effort was made to minimize the free space near the detector. In addition, the passive shielding was enclosed in an aluminum box flushed with radon-free air (<18 mBq/m³) delivered by a radon-free factory installed in the Modane Underground Laboratory [21].

The electronics consisted of currently available spectrometric amplifiers and an 8192 channel ADC. The energy calibration was adjusted to cover the energy range from 50 keV to 3.5 MeV, and the energy resolution was 2.0 keV for the 1332-keV line of ⁶⁰Co. The electronics were stable during the experiment because of the constant conditions in the laboratory (temperature of $\approx 23^{\circ}$ C, hygrometric degree of $\approx 50\%$). A daily check of the apparatus ensured that the counting rate was statistically constant.

The enriched tin sample, disk shaped (the diameter was 67 mm, the height was 2.2 mm), was placed on the endcap

of the HPGe detector. The sample mass was 53.355 g. Taking into account the enrichment of 94.32%, in total 50.3 g of 112 Sn was exposed. The duration of the measurement was 1885.8 h.

The sample was found to have a cosmogenic isotope, ¹¹³Sn ($T_{1/2} = 115.09$ d), with an average activity of (18.8 ± 1.0) mBq/kg. The natural radioactivities had limits that were <3.0 mBq/kg of ²²⁶Ra, <4.6 mBq/kg of ²²⁸Th, <27.2 mBq/kg of ⁴⁰K, and <1.2 mBq/kg of ¹³⁷Cs.

The search for different $\beta^+\text{EC}$ and ECEC processes in ¹¹²Sn were carried out using the germanium detector to look for γ -ray lines corresponding to these processes. The decay scheme for the triplet ¹¹²Sn-¹¹²In-¹¹²Cd is shown in Fig. 1 [22]. The ΔM (difference of parent and daughter atomic masses) value of the transition is 1919.5. $\pm 4.8 \text{ keV}$ [23]. The following decay processes are possible:

$$e_b^- + (A, Z) \to (A, Z - 2) + e^+ + X \quad (\beta^+ EC; 0\nu)$$
(1)

$$+ (A, Z) \rightarrow (A, Z - Z) + e$$
$$+ 2\nu + X \quad (\beta^+ EC; 2\nu) \tag{2}$$

$$2e_{b}^{-} + (A, Z) \rightarrow (A, Z - 2) + 2X \quad (ECEC; 0\nu)$$
 (3)

$$2e_{b}^{-} + (A, Z) \rightarrow (A, Z - 2) + 2\nu + 2X \quad (ECEC; 2\nu), \quad (4)$$

where e_b is an atomic electron and X represents x rays or Auger electrons. Introduced here is the notation Q', which is the effective Q value defined as $Q' = \Delta M - \epsilon_1 - \epsilon_2$ for the ECEC transition and $Q' = \Delta M - \epsilon_1 - 2m_ec^2$ for the β^+ EC process; ϵ_i is the electron binding energy of a daughter nuclide. For ¹¹²Cd, ε is equal to 26.7 keV for the K shell and 4.01, 3.72, and 3.54 keV for the L shell (2s, $2p_{1/2}$, and $2p_{3/2}$ levels) [24]. In the case of the L shell the resolution of the HPGe detector prohibits separation of the lines so we center the study on the 3.72 keV line.

Investigations were made of the β^+ EC transitions to the ground and the 2_1^+ excited states. Additionally, the ECEC transitions to the ground state and six excited states $(2_1^+, 0_1^+, 2_2^+, 0_2^+, 2_3^+, \text{ and } 0_3^+)$ were investigated.

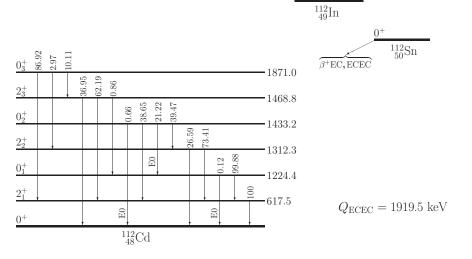
The γ -ray spectra of selected energy ranges are shown in Figs. 2–4. These spectra correspond to regions of interest for the different decay modes of ¹¹²Sn.

percentages.

FIG. 1. Decay scheme of ¹¹²Sn. Only the

investigated levels associated with γ rays are

shown. Transition probabilities are given in



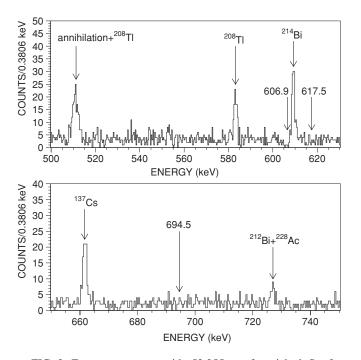


FIG. 2. Energy spectrum with 53.355 g of enriched Sn for 1885.8 h of measurement in the ranges investigated (500–630 and 650–750 keV).

A. ECEC transitions

The ECEC($0\nu + 2\nu$) transition to the excited states of ¹¹²Cd is accompanied with γ quanta with different energies (see decay scheme in Fig. 1). These γ quanta were used in the search. The approach is not sensitive to ECEC(2ν) to the

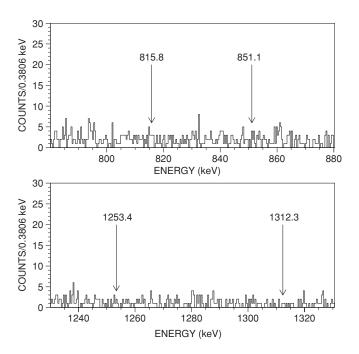


FIG. 3. Energy spectrum with 53.355 g of enriched Sn for 1885.8 h of measurement in the ranges investigated (780–880 and 1230–1330 keV).

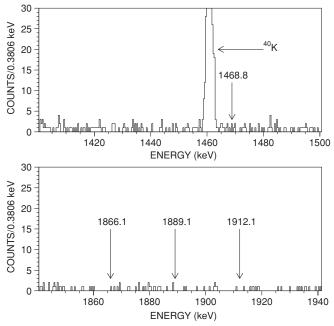


FIG. 4. Energy spectrum with 53.355 g of enriched Sn for 1885.8 h of measurement in the ranges investigated (1400–1500 and 1840–1940 keV).

ground state because x rays are absorbed in the sample and cannot reach the sensitive volume of the HPGe detector.

The ECEC(0ν) transition to the ground state of the daughter nuclei was considered for three different electron capture cases:

- (i) Two electrons were captured from the *L* shell. In this case, Q' was equal to 1912.1 ± 4.8 keV and the transition was accompanied by a bremsstrahlung γ quantum with an energy of ~1912.1 keV.
- (ii) One electron was captured from the K shell and another from the L shell. In this case, Q' was equal to 1889.1 ± 4.8 keV and the transition was accompanied by a bremsstrahlung γ quantum with an energy of ~1889.1 keV.
- (iii) Two electrons were captured from the *K* shell. In this case, Q' was equal to 1866.1 \pm 4.8 keV and the transition was accompanied by a γ quantum with an energy of ~1866.1 keV. In fact this transition was strongly suppressed because of momentum conservation. So in this case the more probable outcome is the emission of an e^+e^- pair [25] that gives two annihilation γ quanta with an energy of 511 keV.

The Bayesian approach [26] was used to estimate limits on transitions of ¹¹²Sn to the ground and excited states of ¹¹²Cd. To construct the likelihood function, every bin of the spectrum is assumed to have a Poisson distribution with its mean μ_i and the number of events equal to the content of the *i*th bin. The mean can be written in the general form

$$\mu_i = N \sum_m \varepsilon_m a_{mi} + \sum_k P_k a_{ki} + b_i.$$
⁽⁵⁾

The first term in Eq. (5) describes the contribution of the investigated process that may have a few γ lines contributing appreciably to the *i*th bin. The parameter N is the number of decays, ε_m is the detection efficiency of the *m*th γ line, and a_{mi} is the contribution of the *m*th line to the *i*th bin. For low-background measurements a γ line may be taken to have a Gaussian shape. The second term gives contributions of background γ lines. Here P_k is the area of the kth γ line and a_{ki} is its contribution to the *i*th bin. The third term represents the so-called "continuous background" (b_i) , which has been selected as a straight-line fit after rejecting all peaks in the region of interest. We have selected this region as the peak to be investigated ± 30 standard deviations (≈ 20 keV). The likelihood function is the product of probabilities for selected bins. Normalizing over the parameter N gives the probability density function for N, which is used to calculate limits for N. To take into account errors in the γ -line shape parameters, peak areas, and other factors, one should multiply the likelihood function by the error probability distributions for these values and integrate to provide the average probability density function for N.

In the case of the ECEC(0ν) transition to the ground state of ¹¹²Cd there is a large uncertainty in the energy of the bremsstrahlung γ quantum because of a poor accuracy in ΔM (±4.8 keV). Thus the position of the peak was varied in the region of the uncertainty and the most conservative value of the limit for the half-life was selected.

The photon detection efficiency for each investigated process has been computed with the CERN Monte Carlo code GEANT 3.21. Special calibration measurements with radioactive sources and powders containing well-known ²²⁶Ra activities confirmed that the accuracy of these efficiencies is about 10%.

The final results are presented in Table I. The fourth column shows the best previous experimental results from Ref. [18] for comparison. In the last column, the theoretical estimations for $ECEC(2\nu)$ transitions obtained under the assumption of single intermediate nuclear state dominance are also presented [27].

Concerning the ECEC(0ν) processes, the plan is to observe a resonant transition to the 1871.0 keV excited state of ¹¹²Cd. In this case we look for two peaks, at 617.5 and 1253.4 keV. In fact, the experimental spectrum has no extra events in the energy range of interest. The conservative approach gives the limit $T_{1/2} > 4.7 \times 10^{20}$ yr at the 90% C.L.

B. β^+ EC transitions

The $\beta^+\text{EC}(0\nu + 2\nu)$ transition to the ground state is accompanied by two annihilation γ quanta with an energy of 511 keV. These γ quanta were used to search for this transition. In the case of the $\beta^+\text{EC}(0\nu + 2\nu)$ transition to the 2_1^+ excited state the 617.4 keV γ quantum was also detected. To obtain limits on these transitions the analysis described in Sec. II A, was used. Again the photon detection efficiencies for each investigated process was computed with the CERN Monte Carlo code GEANT 3.21 and are presented in Table I. The last two columns of the table show the best previous results and theoretical predictions for comparison.

III. DISCUSSION

Limits obtained for the β^+ EC and ECEC processes in ¹¹²Sn are on the level of ~(0.56–8.7) × 10²⁰ yr or ~2–5 times better than the best previous result [18] (see Table I). As one can see from Table I the theoretical predictions for 2 ν transitions are much higher than the measured limits. The sensitivity of such experiments can still be increased with the experimental possibilities being the following:

- (i) Given 1 kg of enriched ¹¹²Sn in the setup described in Sec. II, the sensitivity after 1 yr of measurement will be $\sim 10^{22}$ yr.
- (ii) With 200 kg of enriched ¹¹²Sn, using an installation such as GERDA [28] or Majorana [29,30], where 500–1000 kg of low-background HPGe detectors are planned, is a possibility. Placing ~1 kg of very pure ¹¹²Sn around each of the ~200 HPGe crystals both ⁷⁶Ge and ¹¹²Sn will be investigated at the same time. The sensitivity after 10 yr of measurement may reach ~10²⁶ yr. Thus there is a chance of detecting the β^+ EC(2 ν) transition of ¹¹²Sn to the ground state and the ECEC(2 ν) transition to the 0⁺₁ excited state (see theoretical predictions in Table I).

In the case of the ECEC(0ν) transition to the 0^+_3 (1871.0 keV) excited state of ¹¹²Cd no extra events were detected. So the search for this process continues into the future. Note that the ECEC(2ν) transition to the 0^+_3 excited state is strongly suppressed because of the very small phase space volume. In contrast, the probability of the 0ν transition should be strongly enhanced if the resonance condition is realized. In Refs. [14,15] the "increasing factor" was estimated as $\sim 10^6$ and can be even higher. Then if the "positive" effect is observed in future experiments it is the ECEC(0ν) process. This will mean that lepton number is violated and the neutrino is a Majorana particle. To extract the $\langle m_{\nu} \rangle$ value one must know the nuclear matrix element for this transition and therefore the exact value of ΔM (see Refs. [14,15]). The necessary accuracy for ΔM is better than 1 keV and this is a realistic task (in Ref. [31] the $Q_{\beta\beta}(^{130}\text{Te})$ was measured with an accuracy of 13 eV).

Two different descriptions for the resonance were discussed in the past. In Ref. [14] the resonance condition is realized when Q' is close to zero. They treat the process as (1S, 1S)double electron capture and Q' is equal to -4.9 ± 4.8 keV $(1\sigma \text{ error})$. Thus there is a probability that Q' is less than 1 keV. In this case one has a few daughter-nucleus γ rays (see Fig. 1) and two Cd K x rays, one of which may have its energy shifted by the mismatch in energies between the parent atom and the almost degenerate virtual daughter state. In Refs. [15,32] the decay is treated as (1S, 2P) double electron capture with irradiation of an internal bremsstrahlung photon. The Q' value (energy of the bremsstrahlung photon) is $18.1 \pm$ 4.8 keV. The resonance condition for the transition is realized when $E_{\text{brems}} = Q_{\text{res}} = |E(1S, Z - 2) - E(2P, Z - 2)|;$ i.e., when the bremsstrahlung photon energy becomes comparable to the 2P-1S atomic level difference in the final atom (23 keV). The same effect was theoretically predicted and then experimentally confirmed for single electron capture

Transition	Energy of γ rays keV (Efficiency)	$T_{1/2}^{\exp}$, 10 ²⁰ yr (C.L. 90%)		$T_{1/2}^{\text{th}}(2\nu) (\text{yr}) [27]$
		Present work	Previous work [18]	
β^+ EC(0 ν + 2 ν); g.s.	511.0 (15.2%)	0.56	0.12	3.8×10^{24}
$\beta^{+}\text{EC}(0\nu + 2\nu); 2_{1}^{+}$	617.5 (3.92%)	2.79	0.94	2.3×10^{32}
ECEC(0ν)L ¹ L ² ; g.s.	1912.1 (3.32%)	4.10	1.3	
ECEC(0ν)K ¹ L ² ; g.s.	1889.1 (3.35%)	3.55	1.8	
ECEC(0ν)K ¹ K ² ; g.s.	1866.1 (3.38%)	3.97	1.3	
	511.0 (15.2%)	0.59 ^a	0.12 ^a	
ECEC(0ν); 2_1^+	617.5 (5.53%)	3.93	1.1	
$\text{ECEC}(0\nu); 0_1^+$	606.9 (4.29%)	6.87	1.2	
	617.5 (4.25%)			
ECEC(0ν); 2_2^+	617.5 (3.11%)	3.45	0.89	
	694.9 (2.90%)			
	1312.3 (1.15%)			
ECEC(0ν); 0_2^+	617.5 (3.69%)	2.68	1.6	
	694.9 (1.07%)			
ECEC(0ν); 2_3^+	617.5 (2.64%)	2.64	0.93	
	851.1 (2.09%)			
	1468.8 (1.34%)			
ECEC(0ν); 0_3^+	617.5 (5.09%)	4.66	0.92	
	1253.4 (3.01%)			
ECEC(2ν); 2_1^+	617.5 (6.81%)	4.84	1.2	4.9×10^{28}
$\text{ECEC}(2\nu); 0_1^+$	606.9 (5.42%)	8.67	1.4	7.4×10^{24}
	617.5 (5.35%)			
ECEC(2ν); 2_2^+	617.5 (3.96%)	4.39	1.0	1.9×10^{32}
	694.9 (3.68%)			
	1312.3 (1.47%)			
ECEC(2ν); 0_2^+	617.5 (4.72%)	3.43	1.8	
	694.9 (1.37%)			
ECEC(2 ν); 2 ⁺ ₃	617.5 (3.37%)	3.40	1.0	6.2×10^{31}
	851.1 (2.72%)			
	1468.8 (1.74%)			
ECEC(2ν); 0_3^+	617.5 (5.09%)	4.66	0.92	5.4×10^{34}
	1253.4 (3.01%)			

TABLE I. The experimental limits and theoretical predictions for the β^+ EC and ECEC processes in ¹¹²Sn.

^aFor transition with irradiation of the e^+e^- pair—see text.

(see Discussion in Ref. [32]). It is anticipated, taking into account uncertainties in the Q' value, that the real Q' value is equal to 23 keV with an accuracy of better then 1 keV and the resonance condition is realized. There are a few daughternucleus γ rays (see scheme in Fig. 1), one Cd K x-ray, and a bremsstrahlung photon with energy $\sim K_{\alpha}$. The bremsstrahlung photon may have its energy shifted by the mismatch in energy between the parent atom and the almost degenerate virtual daughter state.

Finally, both approaches predict the same experimental signature for this transition and need to know with better accuracy the value of ΔM to be sure that the resonance condition is really valid. New theoretical investigations of this transition are needed.

IV. CONCLUSION

New limits on β^+ EC and ECEC processes in ¹¹²Sn have been obtained using a 380 cm³ HPGe detector and an external source consisting of 53.355 g enriched tin. In addition, it has been demonstrated that, in future larger-scale experiments, the sensitivity to the ECEC(0ν) processes for ¹¹²Sn can reach the order of 10^{26} yr. Under resonant conditions this decay will be competitive with $0\nu\beta\beta$ decay.

After submission of this article, we became aware of accurate ΔM value measurements for ¹¹²Sn and ¹¹²Cd ($\Delta M =$ 1919.82 ± 0.16 keV [33]). This result disfavors the strong enhancement scenario for the ECEC(0 ν) process to the 0⁺₃ excited state in ¹¹²Cd.

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