Improved limits on β^+EC and ECEC processes in ^{112}Sn

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Limits on β^+EC and ECEC processes in ^{112}Sn have been obtained using a 380 cm^3 HPGe detector and an external source consisting of 100 g enriched tin (94.32% of ^{112}Sn). A limit with 90% C.L. on the ^{112}Sn half-life of 1.3×10^{21} yr for the ECEC(0ν) transition to the 0_3^+ excited state in ^{112}Cd (1871 keV) has been established. This transition has been discussed in the context of a possible enhancement of the decay rate. The limits on other β^+EC and ECEC processes in ^{112}Sn have also been obtained on the level of (0.1–1.6) $\times 10^{21}$ yr at the 90% C.L.

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I. INTRODUCTION

During the last few years, interest in the $\beta^+\beta^+$, β^+EC, and ECEC processes has greatly increased. For the first time a positive result has been obtained in a geochemical experiment with ^{130}Ba, where the ECEC(2ν) process has been detected with a half-life of $(2.2 \pm 0.5) \times 10^{21}$ yr [1]. Recently new strong limits on the ECEC(2ν) process in the promising candidate isotopes (^{78}Kr and ^{106}Cd) have been established (2.4×10^{21} yr [2] and 4.1×10^{20} yr [3], respectively). Also $\beta^+\beta^+$, β^+EC, and ECEC processes in ^{120}Te [4,5], ^{74}Se [6], ^{64}Zn [7–9], ^{108}Cd [10], ^{136}Ce [11], ^{180}W [9], ^{96}Ru [12], and ^{112}Sn [8,13–17] have been investigated. Among the recent papers, there are a few new theoretical papers with half-life estimations [18–26]. Nevertheless, the $\beta^+\beta^+$, β^+EC, and ECEC processes have not been investigated very well theoretically or experimentally. One can imagine some unexpected results here, because of which any further improvements in experimental sensitivity for such transitions has definite merit.

In Ref. [27] it was mentioned that in the case of ECEC(0ν) transition, a resonance condition can exist for transition to the "right energy" of the excited level in the daughter nucleus, where the decay energy is close to zero. In 1982 the same idea was proposed for the transition to the ground state [28]. In 1983 this possibility was discussed for the transition ^{112}Sn to ^{112}Cd (0$^+$; 1871 keV) [29]. In 2004 the idea was reanalyzed in Ref. [30] and new resonance conditions for the decay were formulated. The possible enhancement of the transition rate was estimated to be a factor of $\sim 10^6$ [29,30]. 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, ^{130}Ba, ^{136}Ce, and ^{162}Er) of daughter nuclei exist [30–32]. The precision needed to realize resonance condition is well below 1 keV. Unfortunately, for all the above mentioned isotopes the accuracy is mainly on the level of 2–13 keV. To select the best candidate from the above list, one needs to know the atomic mass difference with an accuracy better than 1 keV. In fact, it is possible to know these values with much better accuracy and recently the atomic-mass difference between ^{112}Sn and ^{112}Cd has been measured with an accuracy of 0.16 keV [37] and between ^{74}Se and ^{74}Ge with an accuracy of 0.007 [33] and 0.049 keV [34] (unfortunately these new measurements indicate that the strongly enhancement scenario for this transition in ^{112}Sn and ^{74}Se is disfavored). The experimental search for such a resonance transition in ^{74}Se to ^{74}Ge (2$^+$; 1206.9 keV) was performed yielding a limit $T_{1/2} > 5.5 \times 10^{18}$ yr [6]. Recently the limits on the level of 1.6×10^{20}, $(0.5–1.3) \times 10^{19}$, and $\sim(2–4) \times 10^{15}$ yr for the resonant neutrinoless transitions in ^{106}Cd [3], ^{96}Ru [12], and ^{136}Ce [11] were obtained. Resonance transition in ^{112}Sn has been investigated by three different experimental groups using natural and enriched samples of tin [13–17]. The stronger limit of $T_{1/2} > 4.7 \times 10^{20}$ yr was obtained for the transition to the 0$^+$ state at 1871 keV with the 53 g enriched tin sample [17].

In this article the results of an experimental investigation of the β^+EC and ECEC processes in ^{112}Sn using 100 g enriched tin sample are presented.

II. EXPERIMENT

The experiment has been performed in the Modane Underground Laboratory at a depth of 4800 m water equivalent (w.e.). The measurements have been done using a 380 cm^3 low-background HPGe detector and enriched tin sample.

The HPGe spectrometer is a *p*-type crystal with the cryostat, endcap, and majority of the mechanical components made of a very pure Al-Si alloy. The cryostat has a *J*-type geometry for shielding the crystal from radioactive impurities in the dewar. The passive shielding consisted of 4 cm of Roman-era lead and 3–10 cm of OFHC copper inside 15 cm of ordinary lead. To remove ^{222}Rn 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 (<10 mBq/kg) delivered by a radon-free factory installed in the Modane Underground Laboratory.

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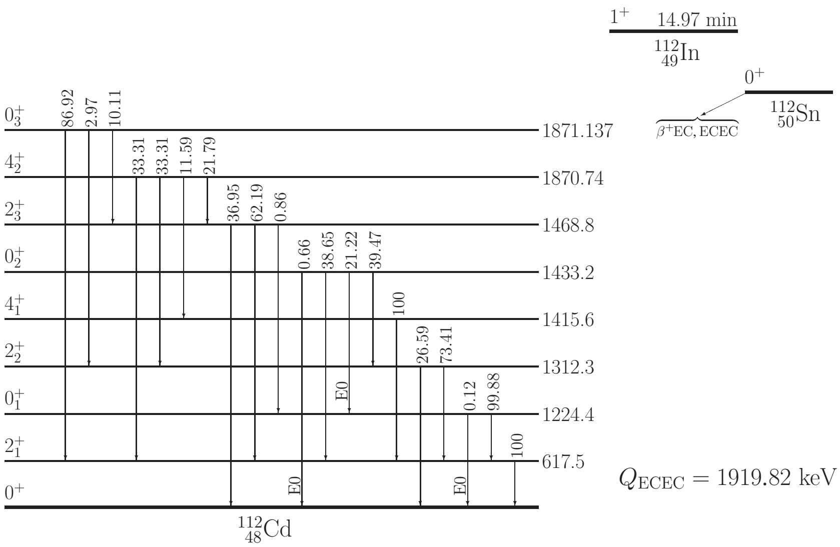

FIG. 1. Decay scheme of ^{112}Sn. Only the investigated levels associated with γ rays are shown. Transition probabilities are given in percents.

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 ^{60}Co. The electronics were stable during the experiment due to the constant conditions in the laboratory (temperature of $\approx 23\,^\circ$C, hygrometric degree of $\approx 50\%$). A daily check on the apparatus assured that the counting rate was statistically constant.

The two enriched disk shaped tin samples (the diameters are 67 and 61 mm, the heights are 2.2 and 2.3 mm) were placed on the endcap of the HPGe detector. The sample mass was 100 g. Taking into account the enrichment of 94.3%, all 94.3 g of ^{112}Sn was exposed. The duration of the measurement was 3175.23 h.

The sample was found to have a cosmogenic isotope, ^{113}Sn ($T_{1/2} = 115.09$ days), with average activity of (11.6 ± 0.5) mBq/kg. The main natural radioactivities have only limits which are <1.0 mBq/kg of ^{226}Ra, <1.8 mBq/kg of ^{228}Th, and <1.2 mBq/kg of ^{137}Cs. For ^{40}K an activity (40 ± 7) mBq/kg has been obtained.

The search for different β^+EC and ECEC processes in ^{112}Sn has been carried out using the germanium detector for looking γ-ray lines corresponding to these processes. The decay scheme for the triplet ^{112}Sn-^{112}In-^{112}Cd is shown in Fig. 1 [35]. Energies of 0_3^+ and 4_2^+ excited states are taken from Ref. [36]. The ΔM (difference of parent and daughter atomic masses) value of the transition has been recently measured as 1919.82 ± 0.16 keV [37] (the best previously measured value was $1919.5. \pm 4.8$ keV [38]). The following decay processes are possible:

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

$$e_b^- + (A, Z) \to (A, Z-2) + e^+ + 2\nu + X \quad (\beta^+\text{EC}; 2\nu), \qquad (2)$$

$$2e_b^- + (A, Z) \to (A, Z-2) + 2X \qquad (\text{ECEC}; 0\nu), \qquad (3)$$

$$2e_b^- + (A, Z) \to (A, Z-2) + 2\nu + 2X \quad (\text{ECEC}; 2\nu), \qquad (4)$$

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 $Q' = \Delta\text{M} - \epsilon_1 - \epsilon_2$ for the ECEC transition and $Q' = \Delta\text{M} - \epsilon_1 - 2m_ec^2$ for the β^+EC process; ϵ_i is the electron binding energy of a daughter nuclide. For ^{112}Cd, ϵ is equal to 26.7 keV for the K shell and 4.01, 3.72, and 3.54 keV for $2s$, $2p_{1/2}$, and $2p_{3/2}$ L shell levels, respectively. In the case of the L shell the resolution of the HPGe detector prohibits separation of the lines so we center our present study on the 3.72 keV line.

Investigations have been made of the β^+EC transitions to the ground and the 2_1^+ excited states. Additionally, the ECEC transitions to the ground state and eight excited states (2_1^+, 0_1^+, 2_2^+, 4_1^+, 0_2^+, 2_3^+, 4_2^+, and 0_3^+) have been investigated.

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 ^{112}Sn.

A. ECEC transitions

The ECEC(0ν) transition to the ground state of the daughter nuclei is accompanied by irradiation of a bremsstrahlung γ quantum with energy Q'. This bremsstrahlung γ quantum is used to detect the ECEC(0ν) transition. There is no bremsstrahlung γ quantum in the case of ECEC(2ν) transition and our method is not sensitive to this process to the ground state because x rays (which are irradiated only) are absorbed in the sample and cannot reach the sensitive volume of the HPGe detector.

The ECEC($0\nu + 2\nu$) transition to the excited states of ^{112}Cd is accompanied with γ quanta with different energies (see decay scheme in Fig. 1). These γ quanta are used in the search. In addition, the ECEC(0ν) transition is accompanied by irradiation of bremsstrahlung γ quantum with energy $Q' - E_i$ (E_i is the energy of an excited state). Most of the ECEC(0ν) decays to excited states suffer a modest decrease in efficiency relative to the equivalent ECEC(2ν) decays because of the potential overlap in the detector of the bremsstrahlung photon with the deexcitation γ, which leads to separate limits for the

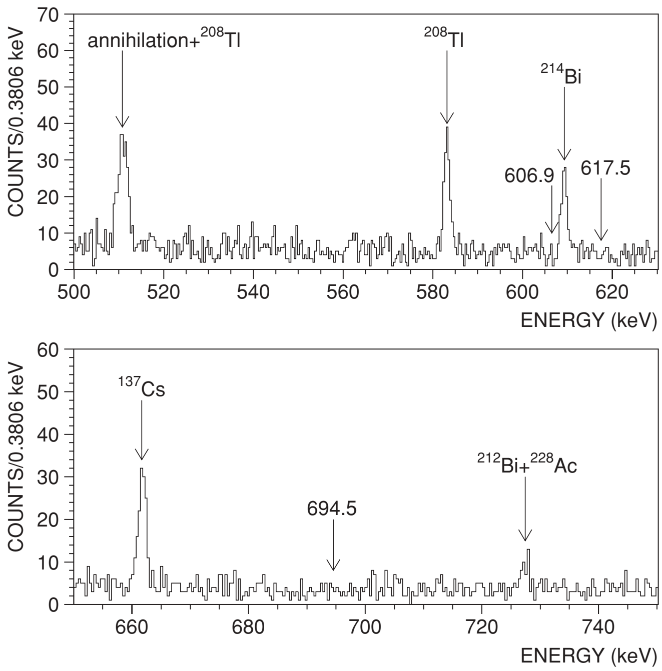

FIG. 2. Energy spectrum with 100 g of enriched Sn for 3175.23 h of measurement in investigated energy ranges [(500–630) and (650–750) keV].

0ν and 2ν modes. In the case of ECEC(0ν) decays to the 4_2^+ and 0_3^+ states the bremsstrahlung photon is too low in energy and cannot be detected by the HPGe detector, that leads to the same limit for 0ν and 2ν modes.

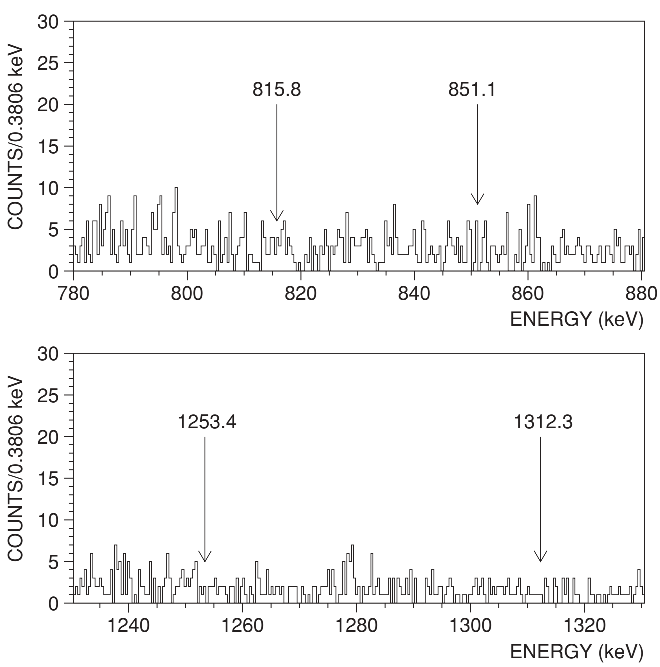

FIG. 3. Energy spectrum with 100 g of enriched Sn for 3175.23 h of measurement in investigated energy ranges [(780–880) and (1230–1330) keV].

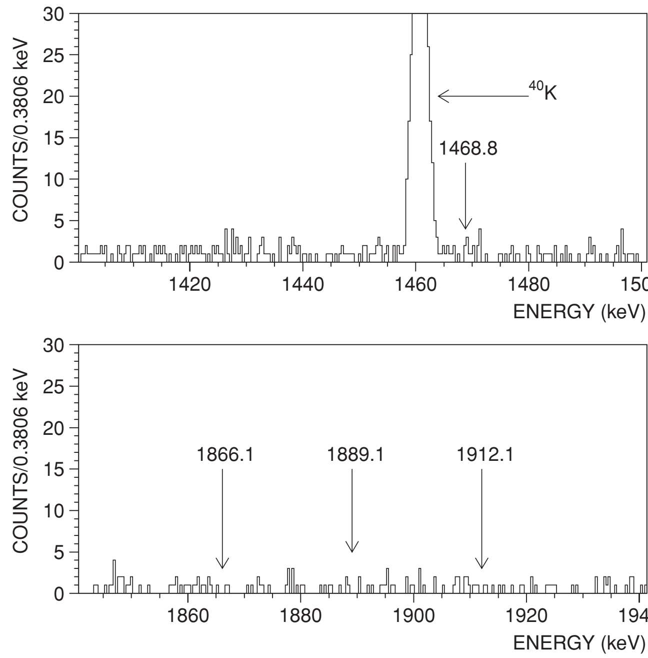

FIG. 4. Energy spectrum with 100 g of enriched Sn for 3175.23 h of measurement in investigated energy ranges [(1400–1500) and (1840–1940) keV].

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

(i) Two electrons are captured from the L shell. In this case Q' is equal to 1912.38 ± 0.16 keV and the transition is accompanied by a bremsstrahlung γ quantum with an energy $\sim$1912.4 keV.
(ii) One electron is captured from the K shell and another from the L shell. In this case Q' is equal to 1889.4 ± 0.16 keV and the transition is accompanied by a bremsstrahlung γ quantum with an energy $\sim$1889.4 keV.
(iii) Two electrons are captured from the K shell. In this case Q' is equal to 1866.42 ± 0.16 keV and the transition is accompanied by γ quantum with an energy $\sim$1866.4 keV. In fact this transition is strongly suppressed because of momentum conservation. So in this case the more probable outcome is the emission of e^+e^- pair [39] which gives two annihilation γ quanta with an energy of 511 keV.

The Bayesian approach [40] has been used to estimate limits on transitions of ^{112}Sn to the ground and excited states of ^{112}Cd. 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 ith bin. The mean can be written in the general form

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

The first term in (5) describes the contribution of the investigated process that may have a few γ lines contributing

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

Transition	Energy of γ rays (keV) (Efficiency)	$T_{1/2}^{exp}$ (10^{20} yr) (C.L. 90%)		$T_{1/2}^{th}(2\nu)$ (yr) [18]
		Present work	Previous work [17]	
β^+EC($0\nu + 2\nu$); g.s.	511.0(13.61%)	0.97	0.56	3.8×10^{24}
β^+EC($0\nu + 2\nu$); 2_1^+	617.5(3.67%)	7.02	2.79	2.3×10^{32}
ECEC(0ν) L^1L^2; g.s.	1912.4(3.12%)	6.43	4.10	
ECEC(0ν) K^1L^2; g.s.	1889.4(3.14%)	8.15	3.55	
ECEC(0ν) K^1K^2; g.s.	1866.4(3.18%)	10.63	3.97	
	511.0(13.61%)	0.97[a]	–	
ECEC(0ν); 2_1^+	617.5(5.08%)	9.72	3.93	
ECEC(0ν); 0_1^+	606.9(3.85%)	12.86	6.87	
	617.5(3.85%)			
ECEC(0ν); 2_2^+	617.5(2.85%)	8.89	3.45	
	694.9(2.69%)			
	1312.3(1.06%)			
ECEC(0ν); 4_1^+	617.5(3.90%)	7.46	–	
ECEC(0ν); 0_2^+	617.5(3.41%)	6.86	2.68	
	694.9(0.99%)			
ECEC(0ν); 2_3^+	617.5(2.48%)	6.46	2.64	
	851.1(2.01%)			
	1468.8(1.25%)			
ECEC(0ν); 4_2^+	558.4(1.52%)	11.03	–	
	617.5(3.59%)			
	1253.2(1.24%)			
ECEC(0ν); 0_3^+	617.5(4.61%)	13.43	4.66	
	1253.4(2.83%)			
ECEC(2ν); 2_1^+	617.5(6.24%)	11.94	4.84	4.9×10^{28}
ECEC(2ν); 0_1^+	606.9(4.89%)	16.25	8.67	7.4×10^{24}
	617.5(4.83%)			
ECEC(2ν); 2_2^+	617.5(3.63%)	11.24	4.39	1.9×10^{32}
	694.9(3.39%)			
	1312.3(1.32%)			
ECEC(2ν); 4_1^+	617.5(5.00%)	9.57	–	
ECEC(2ν); 0_2^+	617.5(4.29%)	8.64	3.43	
	694.9(1.25%)			
ECEC(2ν); 2_3^+	617.5(3.11%)	8.19	3.40	6.2×10^{31}
	851.1(2.52%)			
	1468.8(1.59%)			
ECEC(2ν); 4_2^+	558.4(1.52%)	11.03	–	
	617.5(3.59%)			
	1253.2(1.24%)			
ECEC(2ν); 0_3^+	617.5(4.61%)	13.43	4.66	5.4×10^{34}
	1253.4(2.83%)			

[a]For transition with irradiation of e^+e^- pair see the text.

appreciably to the ith bin. The parameter N is the number of decays, ε_m is the detection efficiency of the mth γ line, and a_{mi} is the contribution of the mth line to the ith 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 ith 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 select this region as the peak to be investigated ± 30 standard deviations ($\approx$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.

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

The final results are presented in Table I. In the fourth column are the best previous experimental results from Ref. [13] for comparison. In the last column the theoretical estimations for ECEC(2ν) transitions obtained under the assumption of single intermediate nuclear state dominance are also presented [18].

Concerning the ECEC(0ν) processes, the plan is to observe a resonant transition to the 0^+(1871.137 keV) excited state of ^{112}Cd. The level structure of ^{112}Cd has been recently investigated in Ref. [36]. Using γ-ray coincidence spectroscopy, decays of the 0^+ and 4^+ doublet of levels at 1871 keV have been isolated, and precise level energies have been determind to be 1871.137(72) and 1870.743(54) keV, respectively. In the case of transition to 0^+ excited state 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 under interest. The conservative approach gives the limit $T_{1/2} > 1.3 \times 10^{21}$ yr at the 90% C.L.

B. β^+EC transitions

The β^+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. The measurements with and without the sample gave approximately the same annihilation peak at 511 keV (within errors). So this peak in our spectrum was considered as background. In the case of the β^+EC($0\nu + 2\nu$) transition to the 2^+_1 excited state the 617.4 keV γ quantum was used. To obtain limits on these transitions the analysis described in Sec. III A was applied. 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. It has to be stressed that since the β^+EC(0ν) and β^+EC(2ν) decays have identical experimental signatures then the efficiency of the decays is the same and a combined limit is reported. In the last two columns are the best previous results and theoretical predictions for comparison.

III. DISCUSSION

Limits obtained for the β^+EC and ECEC processes in ^{112}Sn are in the range of $\sim$(0.1–1.6) $\times 10^{21}$ yr or $\sim$1.5–3 times better than the best previous result [17] (see Table I). The limits on ECEC transitions to the 4^+_1 and 4^+_2 excited states (0ν and 2ν modes) have been obtained. The obtained results are one of the best modern results for such transitions. As one can see from Table I, the theoretical predictions for 2ν 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 ^{112}Sn in the setup described in Sec. II, the sensitivity after 1 yr of measurement will be $\sim 10^{22}$ yr.

(ii) Using 200 kg of enriched ^{112}Sn in an installation such as GERDA [41] or MAJORANA [42,43] where 500–1000 kg of low-background HPGe detectors are planned. Placing about 1 kg of very pure ^{112}Sn around each of the HPGe crystals both ^{76}Ge and ^{112}Sn will be investigated at the same time. The sensitivity after 10 years of measurement may reach $\sim 10^{26}$ yr. Thus there is a chance of detecting the β^+EC(2ν) transition of ^{112}Sn to the ground state and ECEC(2ν) 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 (1871.137 keV) excited state of ^{112}Cd, no extra events were detected. 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. [29,30] the "enhancement factor" was estimated as $\sim 10^6$ (if Q' is $\leqslant$20 eV). 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 has to know the nuclear matrix element for this transition and therefore the exact value of ΔM (see [29,30]). The necessary accuracy for ΔM is better than 1 keV and recently the atomic-mass difference between ^{112}Sn and ^{112}Cd has been measured as 1919.82(16) keV [37]. It means that energy of neutrinoless double electron capture (in which both atomic electrons are captured from the K shell) to 0^+(1871.137 keV) excited state of ^{112}Cd is equal to -4.72 ± 0.23 keV and the strong enhancement scenario for the ^{112}Sn decay is disfavored.

IV. CONCLUSION

New limits on β^+EC and ECEC processes in ^{112}Sn have been obtained using a 380 cm^3 HPGe detector and an external source consisting of 100 g enriched ^{112}Sn. A limit with 90% C.L. on the ^{112}Sn half-life of 1.3×10^{21} yr for the ECEC(0ν) transition to the 0^+_3 excited state in ^{112}Cd (1871 keV) has been established. Using new ΔM value it has been noticed that the strong enhancement scenario for this transition in ^{112}Sn decay is disfavored. In addition, it has been demonstrated that in the future the sensitivity of larger scale experiments can reach the order of 10^{26} yr. In this case there is a chance of detecting the β^+EC(2ν) transition of ^{112}Sn to the ground state and ECEC(2ν) transition to the 0^+_1 excited state.

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