Search for Majorana Neutrinos Near the Inverted Mass Hierarchy Region with KamLAND-Zen

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We present an improved search for neutrinoless double-beta (0νββ) decay of 136Xe in the KamLAND-Zen experiment. Owing to purification of the xenon-loaded liquid scintillator, we achieved a significant reduction of the 108mAg contaminant identified in previous searches. Combining the results from the first and second phase, we obtain a lower limit for the 0νββ decay half-life of $T_{1/2}^{0νββ} > 1.07 \times 10^{28}$ yr at 90% C.L., an almost sixfold improvement over previous limits. Using commonly adopted nuclear matrix element calculations, the corresponding upper limits on the effective Majorana neutrino mass are in the range 61–165 meV. For the most optimistic nuclear matrix elements, this limit reaches the bottom of the quasidegenerate neutrino mass region.

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Neutrinoless double-beta (0νββ) decay is an exotic nuclear process predicted by extensions of the Standard Model of particle physics. Observation of this decay demonstrates the nonconservation of lepton number, and proves that neutrinos have a Majorana mass component. In the framework of light Majorana neutrino exchange, its decay rate is proportional to the square of the effective Majorana neutrino mass $<m_{\nu}> = \sum_i |\sum_j U_{e j}^2 m_{\nu i}|$. Recent 0νββ searches [1] involving 76Ge (GERDA [2]) and 136Xe (KamLAND-Zen [3] and EXO-200 [4]) provide upper limits on $<m_{\nu}>$ of ~0.2–0.4 eV using available nuclear matrix element (NME) values from the literature. The sensitivities of these searches correspond to mass scales in the so-called quasidegenerate mass region.

KamLAND-Zen is a double-beta decay experiment that exploits the existing detection infrastructure and radio-purity of KamLAND [5,6]. The KamLAND-Zen detector consists of 13 tons of Xe-loaded liquid scintillator (Xe-LS) contained in a 3.08-m-diameter spherical inner balloon located at the center of the KamLAND detector. The IB is constructed from 25-μm-thick transparent nylon film and is surrounded by 1 kton of liquid scintillator (LS) contained in a 3.08-m-diameter spherical outer balloon. The outer LS acts as an active shield. The scintillation photons are viewed by 1879 photomultiplier tubes (PMTs) mounted on the inner surface of the containment vessel. The Xe-LS located at the center of the KamLAND detector is 76% 136Xe, 24% 134Xe, and 0.02% 134mXe. Other xenon isotopes have negligible presence. The two electrons emitted from 136Xe ββ decay...
produce scintillation light and their summed energy is observed. Hypothetical $0\nu\beta\beta$ decays would produce a peak at the $Q$ value of the decay, distinguishable from $2\nu\beta\beta$ decays that have a continuous spectrum.

In the first phase of KamLAND-Zen (phase I) [3], we obtained a lower limit of $T^{0\nu}_{1/2} > 1.9 \times 10^{25}$ yr (90% C.L.) on the $^{136}\text{Xe}$ $0\nu\beta\beta$ decay half-life. The sensitivity of the phase-I search was limited by the presence of an unexpected background peak, consistent with $^{110m}\text{Ag}$ $\beta^-$ decay ($\tau = 360$ day, $Q = 3.01$ MeV), just above the 2.458 MeV $Q$ value of $^{136}\text{Xe}$ $\beta\beta$ decay. After completing phase I, we embarked on a Xe-LS purification campaign that continued for 18 months. First, we extracted the Xe-LS in small batches from the IB through a teflon tube whose intake was near the bottom of the IB volume. We then isolated and stored the Xe before placing the Xe-depleted LS back in the top of the IB where it was later replaced by a new LS. This new LS was initially purified by water extraction followed by vacuum distillation. The replacement of the Xe-depleted LS was performed in three cycles equivalent to one IB volume exchange for each cycle. The LS was purified by vacuum distillation during each cycle. We also purified a mix of recovered and new Xe through distillation and refining with a heated zirconium getter. Finally, the Xe was dissolved into the purified LS. In December of 2013, we dissolved into the purified LS. In December of 2013, we dissolved into the purified LS. In December of 2013, we dissolved into the purified LS. In December of 2013, we dissolved into the purified LS. In December of 2013, we dissolved into the purified LS. In December of 2013, we dissolved into the purified LS. In December of 2013, we dissolved into the purified LS. In December of 2013, we dissolved into the purified LS.
between $^{214}$Bi and $^{214}$Po sequential decay events from the initial $^{222}$Rn contamination within the Xe-LS.

An enlarged 3.5-m-radius spherical volume was used to study a high statistics sample of muon spallation products and better constrain their background contributions. This included a region outside the IB. We assess a 22% systematic uncertainty on the calculated spallation yields in the Xe-LS, taking account of the observed ($20 \pm 2$)% increase in the spallation-neutron flux in the Xe-LS relative to the outer LS. In the $0\nu\beta\beta$ window, events from $^{10}$C decays ($\beta^+$, $\tau = 27.8$ s, $Q = 3.65$ MeV) dominate the contribution from muon spallation. A triple-coincidence tag of a muon, a neutron identified by neutron-capture $\gamma$ rays, and the $^{10}$C decay [11], reduces the $^{10}$C background with an efficiency of (64 ± 4)%. Post-muon spallation-neutron events are recorded by newly introduced dead-time free electronics. We apply spherical volume cuts ($\Delta R < 1.6$ m) around the reconstructed neutron vertices for 180 s after the preceding muon. We estimate that the remaining $^{10}$C background after cuts is $(1.01 \pm 0.26) \times 10^{-2}$ (ton day)$^{-1}$, where ton is a unit of Xe-LS mass. Other shorter-lived products, e.g., $^6$He and $^{12}$B, are also reduced by the triple-coincidence tag and have a minor contribution to the background. The dead time introduced by all the spallation cuts is 7%. In the Xe-LS, long-lived $^{137}$Xe ($\beta^-$, $\tau = 5.5$ min, $Q = 4.17$ MeV) is a background source produced by neutron capture on $^{136}$Xe. Based on the spallation-neutron rate and the $^{136}$Xe capture cross section [12], the production yield of $^{137}$Xe is estimated to be $(3.9 \pm 2.0) \times 10^{-3}$ (ton day)$^{-1}$, which is consistent with the simulation study in FLUKA [13,14].

We perform the $0\nu\beta\beta$ decay analysis using a 2-m-radius fiducial volume (FV) as described above to utilize the deployed $^{136}$Xe mass. However, the sensitivity is dominated by the innermost 1-m-radius spherical volume due to the background from the IB. The region outside this radius serves to strongly constrain the tails of the IB background extending into the innermost region. Further, anticipating the decay of the $^{110m}$Ag background identified in phase I, we divide the phase-II data set into two equal time periods (period 1 and period 2), each roughly equal to one average lifetime of the $^{110m}$Ag decay rate. Table I lists the number of observed events, and the estimated and best-fit background contributions in the $0\nu\beta\beta$ window within a 1-m-radius spherical volume for each of the two time periods. The fit is described in detail below. We find a precipitous decrease in the event rate in the $0\nu\beta\beta$ window in period 2. The period-2 background components are well constrained near the values listed in Table I with the exception of $^{110m}$Ag. The hypothesis of standard radioactive decay of the $0\nu\beta\beta$ window background with the decay rate of $^{110m}$Ag is disfavored relative to the hypothesis of a faster reduction at 96% C.L. The origin of this apparent reduction of $^{110m}$Ag is unknown, but we speculate that much of it settled to the bottom of the IB where only a small fraction of $^{110m}$Ag decays is reconstructed in the inner Xe-LS volume. In order to allow the $0\nu\beta\beta$ window background the greatest freedom in the fit, the $0\nu\beta\beta$ decay analyses are performed independently for period 1 and period 2.

The $2\nu\beta\beta$ decay rate can, in principle, be estimated from the same analysis used to derive the $0\nu\beta\beta$ decay limits, but the region outside of 1-m radius contributes negligibly to the $2\nu\beta\beta$ decay rate estimate and is dominated by systematic uncertainty arising from the IB background. To obtain a
2νββ decay rate free of such systematic uncertainty, we perform a separate estimate using a likelihood fit to the binned energy spectrum of the selected candidates between 0.5 and 4.8 MeV, limited to a volume within the 1-m-radius spherical fiducial volume (FV0.5 and 4.8 MeV, limited to a volume within the 1-m-radius spherical volume for each of the two time periods).

<table>
<thead>
<tr>
<th>Observed events</th>
<th>Estimated</th>
<th>Best-fit</th>
</tr>
</thead>
<tbody>
<tr>
<td>136Xe 2νββ</td>
<td>...</td>
<td>5.48</td>
</tr>
<tr>
<td>Residual radioactivity in Xe-LS</td>
<td></td>
<td></td>
</tr>
<tr>
<td>214Bi (238U series)</td>
<td>0.23 ± 0.04</td>
<td>0.25</td>
</tr>
<tr>
<td>208Tl (232Th series)</td>
<td>...</td>
<td>0.001</td>
</tr>
<tr>
<td>110mAg</td>
<td>...</td>
<td>8.5</td>
</tr>
<tr>
<td>External (Radioactivity in IB)</td>
<td></td>
<td></td>
</tr>
<tr>
<td>214Bi (238U series)</td>
<td>...</td>
<td>2.56</td>
</tr>
<tr>
<td>208Tl (232Th series)</td>
<td>...</td>
<td>0.02</td>
</tr>
<tr>
<td>110mAg</td>
<td>...</td>
<td>0.003</td>
</tr>
<tr>
<td>Spallation products</td>
<td></td>
<td></td>
</tr>
<tr>
<td>10C</td>
<td>2.7 ± 0.7</td>
<td>3.3</td>
</tr>
<tr>
<td>6He</td>
<td>0.07 ± 0.18</td>
<td>0.08</td>
</tr>
<tr>
<td>13B</td>
<td>0.15 ± 0.04</td>
<td>0.16</td>
</tr>
<tr>
<td>137Xe</td>
<td>0.5 ± 0.2</td>
<td>0.5</td>
</tr>
</tbody>
</table>

The possible background contributions from 110mAg on the IB film are radially attenuated but larger in the lower hemisphere. So we divide the FV into 20 equal-volume bins for each of the upper and lower hemispheres [see Fig. 1(a)]. We perform a simultaneous fit to the energy spectra for all volume bins. The dependence of 214Bi on the IB film is extracted from a fixed energy window dominated by these events. The 214Bi background contribution is then broken into two independent distributions in the upper and lower hemispheres whose normalizations are floated as free parameters. The fit reproduces the energy spectra for each volume bin; Fig. 1(b) shows an example of the energy spectrum in a volume bin with high 214Bi background events around the IB film. The radial dependences of candidate events and best-fit background contributions in the 0νββ window are illustrated in Fig. 1(c).

The possible background contributions from 110mAg are free parameters in the fit. We consider three independent components: 110mAg uniformly dispersed in the Xe-LS volume and on the surfaces of each the lower and upper IB films. We also examined nonuniform 110mAg sources, with different assumed radial dependences, in the Xe-LS but determined that this has little impact on the 0νββ limit.

As described above, the fits are performed independently for period 1 and period 2 in the region 0.8 < E < 4.8 MeV. We found no event excess over the background expectation for both data sets. The 90% C.L. upper limits on the 136Xe 0νββ decay rate are < 5.5 (kton day)−1 and < 3.4 (kton day)−1 for period 1 and period 2, respectively.
To demonstrate the low background levels achieved in the $0\nu\beta\beta$ region, Fig. 2 shows the energy spectra within a 1-m radius, together with the best-fit background composition and the 90% C.L. upper limit for $0\nu\beta\beta$ decays. Combining the results, we obtain a 90% C.L. upper limit of $< 2.4 \text{ (kton day)}^{-1}$, or $T_{1/2}^{0\nu} > 9.2 \times 10^{25}$ yr (90% C.L.). We find that a fit including potential backgrounds from $^{88}Y$, $^{208}Bi$, and $^{60}Co$ [3] does not change the obtained limit. A MC of an ensemble of experiments assuming the best-fit background spectrum without a $0\nu\beta\beta$ signal indicates a sensitivity of $5.6 \times 10^{25}$ yr, and the probability of obtaining a limit stronger than the presented result is 12%. For comparison, the sensitivity of an analysis in which the $^{110m}Ag$ background rates in period 1 and period 2 are constrained to the $^{110m}Ag$ half-life is $4.5 \times 10^{25}$ yr. Combining the phase-I and phase-II results, we obtain $T_{1/2}^{0\nu} > 1.07 \times 10^{26}$ yr (90% C.L.). This corresponds to an almost sixfold improvement over the previous KamLAND-Zen limit using only the phase-I data, owing to a significant reduction of the $^{110m}Ag$ contaminant and the increase in the exposure of $^{136}Xe$.

From the limit on the $^{136}Xe$ $0\nu\beta\beta$ decay half-life, we obtain a 90% C.L. upper limit of $(m_{\beta\beta}) < (61 - 165)$ meV using an improved phase space factor calculation [17,18] and commonly used NME calculations [19–25] assuming the axial coupling constant $g_A \approx 1.27$. Figure 3 illustrates the allowed range of $(m_{\beta\beta})$ as a function of the lightest neutrino mass $m_{\text{lightest}}$ under the assumption that the decay mechanism is dominated by exchange of a pure-Majorana Standard Model neutrino. The shaded regions include the uncertainties in $U_{ei}$ and the neutrino mass splitting, for each hierarchy. Also drawn are the experimental limits from the $0\nu\beta\beta$ decay searches for each nucleus [2,26–28]. The upper limit on $(m_{\beta\beta})$ from KamLAND-Zen is the most stringent, and it also provides the strongest constraint on $m_{\text{lightest}}$ considering extreme cases of the combination of CP phases and the uncertainties from neutrino oscillation parameters [29,30]. We obtain a 90% C.L. upper limit of $m_{\text{lightest}} < (180–480)$ meV.

In conclusion, we have demonstrated effective background reduction in the Xe-loaded liquid scintillator by purification, and enhanced the $0\nu\beta\beta$ decay search sensitivity in KamLAND-Zen. Our search constrains the mass

![FIG. 2. (a) Energy spectrum of selected $\beta\beta$ candidates within a 1-m-radius spherical volume in period 2 drawn together with best-fit backgrounds, the $2\nu\beta\beta$ decay spectrum, and the 90% C.L. upper limit for $0\nu\beta\beta$ decay. [(b) and (c)] Close-up energy spectra for $2.3 < E < 3.0$ MeV in period 1 and period 2, respectively.](image)

![FIG. 3. Effective Majorana neutrino mass $(m_{\beta\beta})$ as a function of the lightest neutrino mass $m_{\text{lightest}}$. The dark shaded regions are the predictions based on best-fit values of neutrino oscillation parameters for the normal hierarchy (NH) and the inverted hierarchy (IH), and the light shaded regions indicate the $3\sigma$ ranges calculated from the oscillation parameter uncertainties [29,30]. The horizontal bands indicate 90% C.L. upper limits on $(m_{\beta\beta})$ with $^{136}Xe$ from KamLAND-Zen (this work), and with other nuclei from Refs. [2,26–28], considering an improved phase space factor calculation [17,18] and commonly used NME calculations [19–25]. The side panel shows the corresponding limits for each nucleus as a function of the mass number.](image)
scale to lie below \( \sim 100 \) meV, and the most advantageous nuclear matrix element calculations indicate an effective Majorana neutrino mass limit near the bottom of the quasidegenerate neutrino mass region. The current KamLAND-Zen search is limited by backgrounds from \(^{214}\text{Bi}\), \(^{110}\text{m}\text{Ag}\), and muon spallation, and partially by the tail of \(2\nu\beta\beta\) decays. In order to improve the search sensitivity, we plan to upgrade the KamLAND-Zen experiment with a larger Xe-LS volume loaded with 800 kg of enriched Xe, corresponding to a twofold increase in \(^{130}\text{Xe}\), contained in a larger balloon with lower radioactive background contaminants. If further radioactive background reduction is achieved, the background will be dominated by muon spallation, which can be further reduced by optimization of the spallation cut criteria. Such an improved search will allow \(m_{\beta\beta}\) to be probed below 50 meV, starting to constrain the inverted mass hierarchy region under the assumption that neutrinos are Majorana particles. The sensitivity of the experiment can be pushed further by improving the energy resolution to minimize the leakage of the \(2\nu\beta\beta\) tail into the \(0\nu\beta\beta\) analysis window. Such improvement is the target of a future detector upgrade.

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