

Improved intensities for the γ transitions with $E_\gamma > 3$ MeV from $^{208}\text{Pb}^*$ G. W. Kim ¹, K. I. Hahn,^{2,3,*} W. G. Kang,¹ Y. D. Kim,^{1,4,5} E. K. Lee,¹ M. H. Lee ^{1,6}, D. S. Leonard,¹ and S. Y. Park^{1,7}¹Center for Underground Physics, Institute for Basic Science (IBS), Daejeon 34126, Korea²Department of Science Education, Ewha Womans University, Seoul 03760, Korea³Center for Exotic Nuclear Studies, Institute for Basic Science (IBS), Daejeon 34126, Korea⁴Department of Physics and Astronomy, Sejong University, Seoul 05006, Korea⁵University of Science and Technology (UST), Daejeon 34113, Korea⁶IBS School, University of Science and Technology (UST), Daejeon 34113, Korea⁷Department of Physics, Ewha Womans University, Seoul 03760, Korea

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^{208}Pb is a doubly magic nucleus which has been studied extensively by many experiments and nuclear models. Its properties, such as excitation energies, spins, and parities of the ground and excited states are well known. However, some γ transitions with $E_\gamma > 3$ MeV from the excited states to the ground state in ^{208}Pb have never been observed. The intensities of these γ transitions are important issues for $0\nu\beta\beta$ decay searches because these transitions can make non-negligible background signals depending on their intensities. We conducted a measurement to search for the 3197.7, 3475.1, and 3708.4 keV transitions, which are emitted from the second, third, and fourth excited states in ^{208}Pb , respectively. A 2 kg ThO_2 sample was measured using a 100% relative efficiency high-purity germanium detector for 39.5 days at Y2L, an underground laboratory in Yangyang, Korea. We improved upper limits for intensities of the 3197.7, 3475.1, and 3708.4 keV γ transitions as $<1.2 \times 10^{-4}\%$, $<1.4 \times 10^{-4}\%$, and $<1.2 \times 10^{-4}\%$, respectively. These upper limits are at least a factor of 19 better than the previous results.

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I. INTRODUCTION

Doubly-magic nuclei, which have proton and neutron numbers both equal to one of the magic numbers, are very interesting nuclei. These nuclei are usually stable and are suitable examples to verify nuclear shell models [1–3]. ^{208}Pb is one of the doubly-magic nuclei, and its properties have provided crucial clues to our understanding of nuclear structure [4,5]. Information for the level structure of ^{208}Pb has been obtained by various types of experiments [6–9]. In particular, γ -ray spectroscopy is widely used to study excited states at the relatively low-energy region below 4 MeV [9–11]. However, several γ emissions with $3 < E_\gamma < 4$ MeV, such as the 3197.7, 3475.1, and 3708.4 keV transitions, have never been observed. The main reasons for the difficulty to observe these γ transitions are that (a) the expected intensities are extremely small and (b) measurement with a high-purity germanium (HPGe) detector may not distinguish between summed signals arising from true coincidence or accidental coincidence and a single γ ray with the same energy.

Apart from the general nuclear structure study, γ transitions from excited states of ^{208}Pb are important issues for certain rare process experiments, such as neutrinoless double beta ($0\nu\beta\beta$) decay searches, because ^{208}Pb is the daughter product of ^{208}Tl decay which is the final process in the ^{232}Th decay chain. ^{208}Tl backgrounds are one of the most significant

limitations on $0\nu\beta\beta$ decay search experiments [12] because the decay has high Q value and can happen naturally in essentially all detector materials. Many $0\nu\beta\beta$ decay search experiments use isotopes with high Q values, such as ^{48}Ca , ^{130}Te , and ^{100}Mo , for their experiments. The AMoRE experiment selected ^{100}Mo as an isotope for the $0\nu\beta\beta$ decay search [13]. The 3.0 MeV Q value of ^{100}Mo is high enough to avoid background signals from most radioactive decays, with the exception of a few decays such as ^{208}Tl . The Q value of ^{208}Tl is 5.0 MeV, and its decay can make background signals at the region of interest (ROI) in $0\nu\beta\beta$ decay search experiments. While its γ emissions, the form of radiation most prone to producing background signals, are mostly below 2614.5 keV, high-energy γ emissions from the excited states of ^{208}Pb should be investigated carefully.

The 3197.7, 3475.1, and 3708.4 keV γ transitions can occur [14], but these emissions have never been observed. Vasil'ev *et al.* performed a measurement using an HPGe detector to constrain intensities for these γ transitions [15]. They reported only upper-limit values for intensities, which are $<0.007\%$, $<0.003\%$, and $<0.004\%$ for the 3197.7, 3475.1, and 3708.4 keV γ transitions, respectively [14,15]. However, these upper-limit values are not small enough to ignore effects of the 3197.7, 3475.1, and 3708.4 keV γ transitions in some of the $0\nu\beta\beta$ decay search experiments. If current upper limits are assumed to be the actual intensities, a Monte Carlo simulation study showed that the 3197.7, 3475.1, and 3708.4 keV γ transitions can produce non-negligible internal background signals in the ROI of the AMoRE experiment

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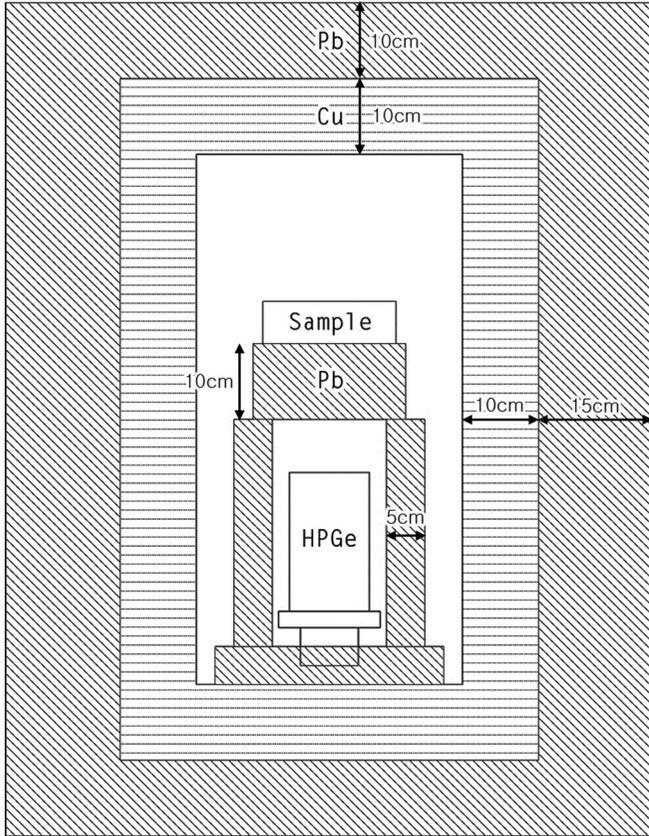


FIG. 1. A schematic of an HPGe detector and shielding structure. The ThO_2 powder sample was placed on top of a 10 cm thick lead block to reduce coincidence summing compared to single- γ transitions with the same energy.

[16]. Hence, tighter constraints on the values of intensities for these γ transitions are necessary to understand their exact effects on backgrounds of $0\nu\beta\beta$ decay search experiments, and particularly on reaching the zero-background level in the AMoRE experiment. We conducted a measurement to obtain improved numbers for constraints on the intensities, compared to the existing results of Vasil'ev *et al.* [15], for the 3197.7, 3475.1, and 3708.4 keV γ transitions from the corresponding excited states of ^{208}Pb .

II. EXPERIMENTAL SETUP

We used a 2 kg ThO_2 powder as a sample source for measuring intensities of the γ transitions from excited states of ^{208}Pb with $E_\gamma > 3$ MeV. The powder was packed in a plastic bag and shaped as a rectangular box with dimensions of 17.5 cm \times 13.0 cm \times 5.6 cm. The radioactivities of the powder were measured to be 28.0(28) kBq/kg for ^{228}Ac and 66.0(42) kBq/kg for ^{228}Th . The measurement was conducted at the Yangyang underground laboratory (Y2L), which is located in an underground tunnel at a depth of 700 m (2000 m.w.e.) in Yangyang, Korea [17,18]. A P-type coaxial HPGe detector with 100% relative detection efficiency was used for this measurement, and the shielding system and experimental setup are shown in Fig. 1. This setup produced

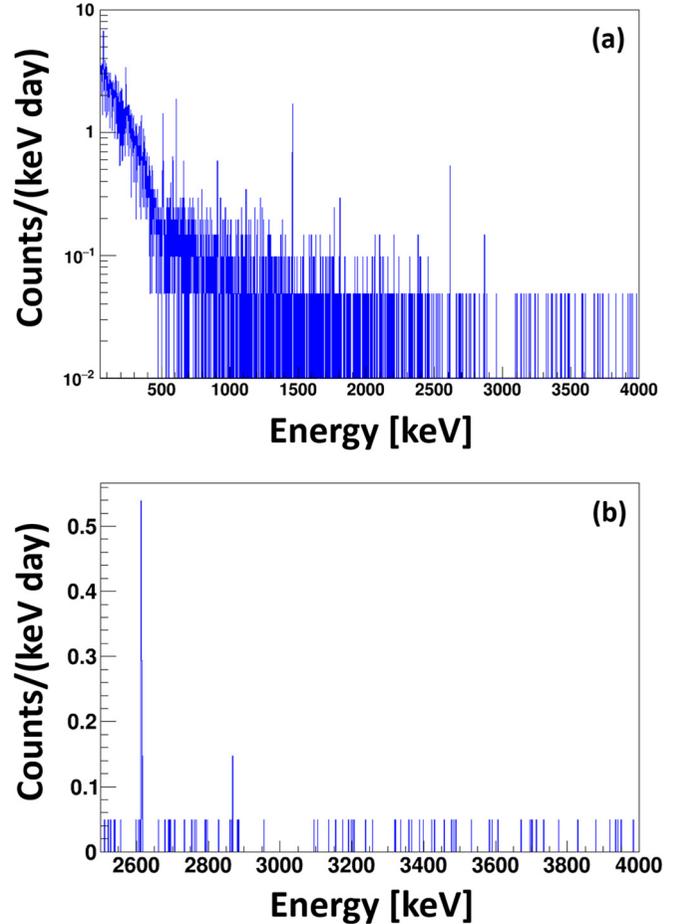


FIG. 2. Background spectrum of the HPGe detector (a) in the energy range from 50 to 4000 keV (logarithmic scale from 10^{-2} for the y axis) and (b) in the 3000 to 4000 keV energy region.

a total observed event rate of about 84 Hz. With our 64 μs analog to digital converter (ADC) window, this produced dead time of about 0.5%.

Figure 2 shows background level of the HPGe detector. The total energy spectrum from 50 to 4000 keV energy range is shown in Fig. 2(a) and the total background count rate in this range is about 0.0080 Hz. Figure 2(b) shows the energy spectrum from 2.5 to 4 MeV and there is no discernible peak other than the 2614.5 keV peak from ^{208}Tl decay. The background count rate from 3 to 4 MeV, the energy region of interest in this measurement, is only 2.38×10^{-5} Hz.

Figure 3 shows a decay scheme of ^{208}Tl to the first four excited states of ^{208}Pb . Except for pileup from random coincidence, the 3197.7 keV peak can be produced in two different ways: one is a single- γ transition with $E_\gamma = 3197.7$ keV and the other is a true coincidence summing of the 583.2 and 2614.5 keV γ transitions. In the same way, both 3475.1 and 3708.4 keV peaks will include single- γ transitions and pileup from true coincidence. γ rays passing through some material are suppressed as follows:

$$I/I_0 = e^{-\mu L}, \quad (1)$$

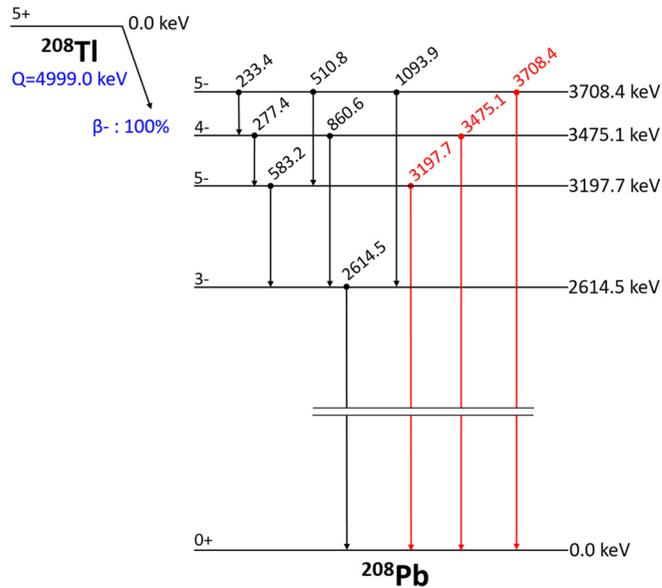


FIG. 3. Decay scheme of ^{208}Tl to the first four excited states of ^{208}Pb

where I_0 is the initial γ intensity, I is the reduced intensity after passing through a material with thickness of L , and μ_l is the attenuation coefficient [19]. In general, the μ_l value for γ rays with high energy is smaller than that for those with low energy [20]. Based on a Monte Carlo study, a 10 cm thick lead block was placed under the bottom of the ThO_2 powder sample. This lead block was expected to reduce the flux of low-energy γ rays incident on the detector relative to the flux from γ transitions with $E_\gamma > 3$ MeV.

We calculated attenuation ratios I/I_0 for γ rays with several energies, 583.2, 860.6, 2614.5, 3197.7, 3475.1, and 3708.1 keV, to see the difference between the single- γ transition and the coincidence summing. The coincidence summing of

the 1093.9 and 2614.5 keV γ transitions was ignored in this study because intensity of the 1093.9 keV transition is only 0.43% compared to other high values such as intensities of the 583.2 keV (85.0%) and 860.6 keV (12.5%) transitions [14]. According to the NIST database [21], the attenuation ratios for 583.2, 860.4, and 2614.5 keV γ rays passing directly through 10 cm of lead are about 1.10×10^{-6} , 1.41×10^{-4} , and 7.86×10^{-3} , respectively. The attenuation ratio for the 3197.7 keV γ ray is 8.73×10^{-3} , and it is 8.85×10^{-3} for both 3475.1 and 3708.4 keV γ rays. The attenuation ratio for the coincidence of 583.2 and 2614.5 keV γ rays can be estimated as the product of the two attenuation ratios, yielding only 8.64×10^{-9} . In the same way, the attenuation ratio for the coincidence of 860.6 and 2614.5 keV γ rays is 1.11×10^{-6} . These calculations show that the experimental setup with a 10 cm lead block can be effective to reduce true coincidence summing compared to the single- γ transitions in the 3197.7, 3475.1, and 3708.4 keV energy regions. In addition, the solid angle factor for our detection system is also squared in the coincidence probability, providing a further improvement.

III. DATA ANALYSIS

Figure 4 shows the γ -energy spectrum of the ThO_2 powder measured for 39.5 days. In the low-energy region of $E_\gamma < 600$ keV, no peaks were found except at 511 keV. The 511 keV peak is most likely from the electron-positron annihilation (510.9 keV) [22], not the 510.8 keV γ ray from the ^{232}Th decay. The spectrum indicates that low-energy γ rays from the sample were suppressed effectively by the 10 cm thick lead block, as expected.

The previous experiment by Vasil'ev *et al.* used 20 g of a thorium powder source [15]. To allow for more shielding and relative suppression of coincidence, we increased the mass of the ThO_2 sample to 2 kg. The larger amount of sample can increase not only the detection possibility of peaks in

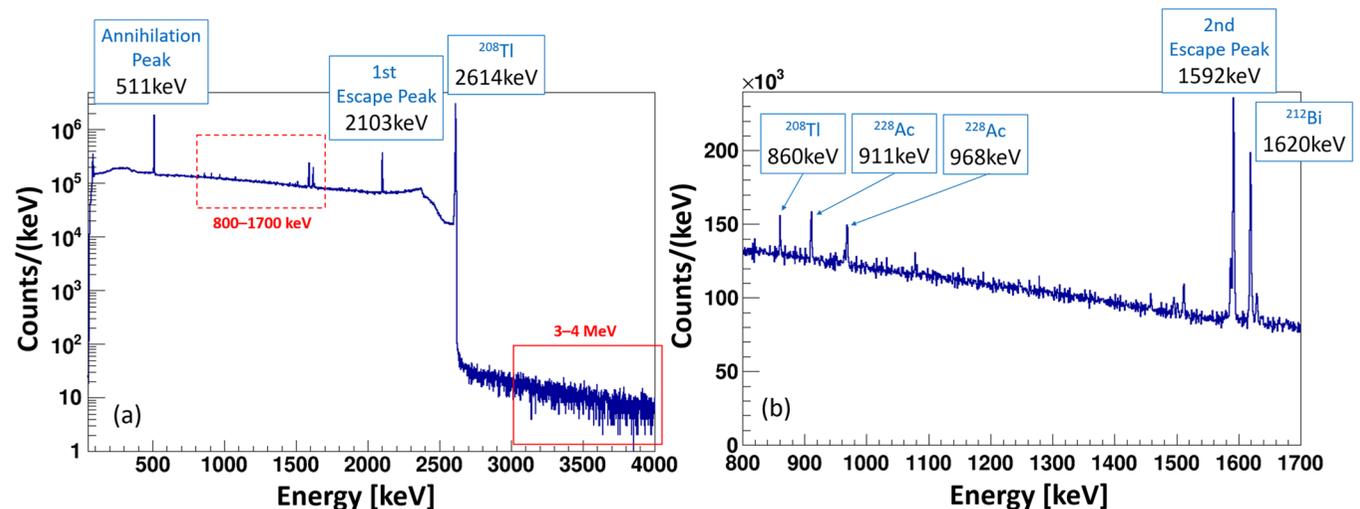


FIG. 4. Energy spectrum of the ThO_2 powder sample for 39.5 days of data taking. (a) The energy spectrum from 0 to 4000 keV (logarithmic scale from 1 for the y axis). Events in the range above 2614 keV are mainly from random pileup. (b) The energy spectrum from 800 to 1700 keV.

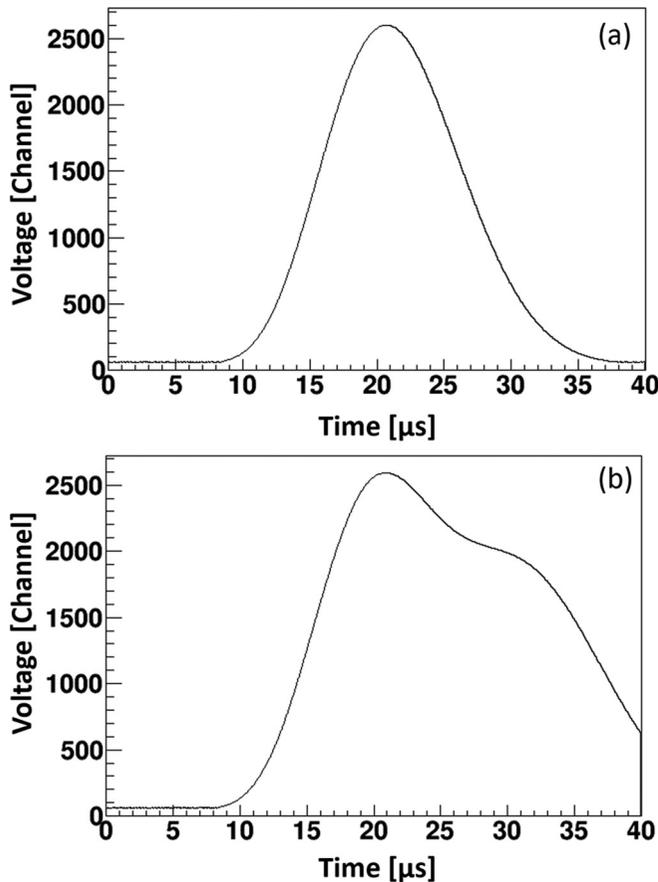


FIG. 5. Comparison of waveforms from (a) a single- γ event and (b) a pileup event. The FWHM values of the two waveforms for similar γ energy are different.

the ROI, but also increase continuum pileup backgrounds. In the obtained data, as expected, events above 3 MeV were dominated by pileup events caused by random coincidence. Since the low-energy peaks have been effectively suppressed, the pileup at the energies of interest is expected to arise from coincidence involving γ rays from the Compton continuum. They do not create false peaks but do limit the source strength that can be used before peak detection sensitivity becomes limited by pileup. In order to further increase the sensitivity, it was necessary to minimize these pileup events.

Single events and pileup events can be distinguished by the full width at half maximum (FWHM) timing values of their waveforms. Figure 5 shows a clear difference between waveforms which have similar maximum voltage values for a single event and a pileup event. The FWHM value of a single event waveform is usually smaller than that of a pileup event. In order to distinguish single events from pileup events, a scatter plot of FWHM of time versus energy was used, as shown in Fig. 6. In the low-energy region ($E_\gamma < 500$ keV), the FWHM values increase from 8.6 to 11.7 μs as the γ energy increases. However, for the energy region above 500 keV, the FWHM values increase slowly as the energy increases. The FWHM values of single events over 2 MeV are converging from 11.7 to 12.1 μs . We used FWHM > 12.1 μs as the cut

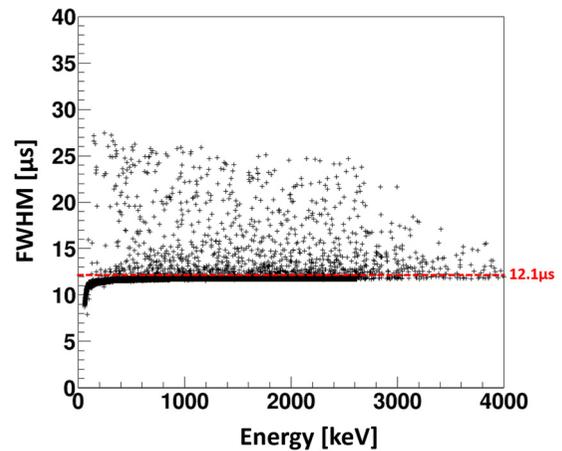


FIG. 6. FWHM of waveform vs energy for 4 hours of data. The events with FWHM > 12.1 μs (above the red line) were considered as pileup events in this study.

condition for the pileup rejection in the $E_\gamma > 3$ MeV region containing the peaks of interest.

The γ events at $E_\gamma = 2614.5$ keV are useful to check the validity of this cut condition, because the 2614.5 keV peak is only produced by single- γ events from the first excited state to the ground state in ^{208}Pb , with background produced by random coincidence with Compton-scattered γ rays. There is no true coincidence summing which has 2614.5 keV energy, and the signal waveforms with 2614.5 keV energy have mostly the same height and FWHM values. Therefore, the 2614.5 keV peak should not be affected significantly by the cut condition. In this measurement, less than 0.02% of the total 2615.5 keV γ events were rejected after applying the cut condition. For the events with $E_\gamma > 3$ MeV, 66% of events were rejected by the cut condition. Figure 7 shows a comparison of two energy spectra with and without the pileup rejection.

Likewise we can check the cut efficiency at the energies of interest using the true coincidence events at those energies. We looked at the cut efficiency using a 0.1 g ThO_2 sample was placed directly on the detector. Out of 897 events observed in

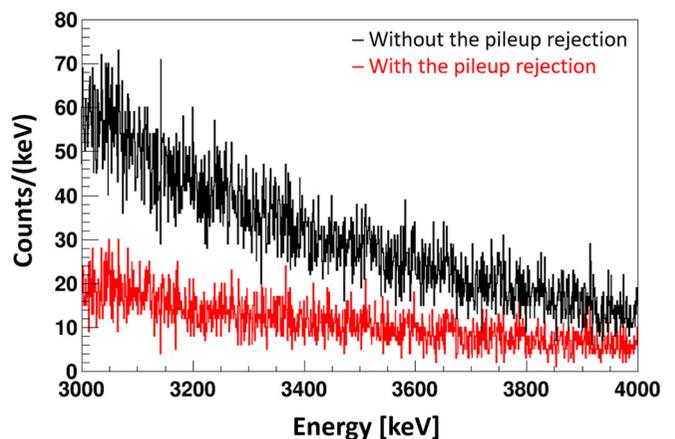


FIG. 7. Energy spectra from 3 to 4 MeV with and without the pileup rejection.

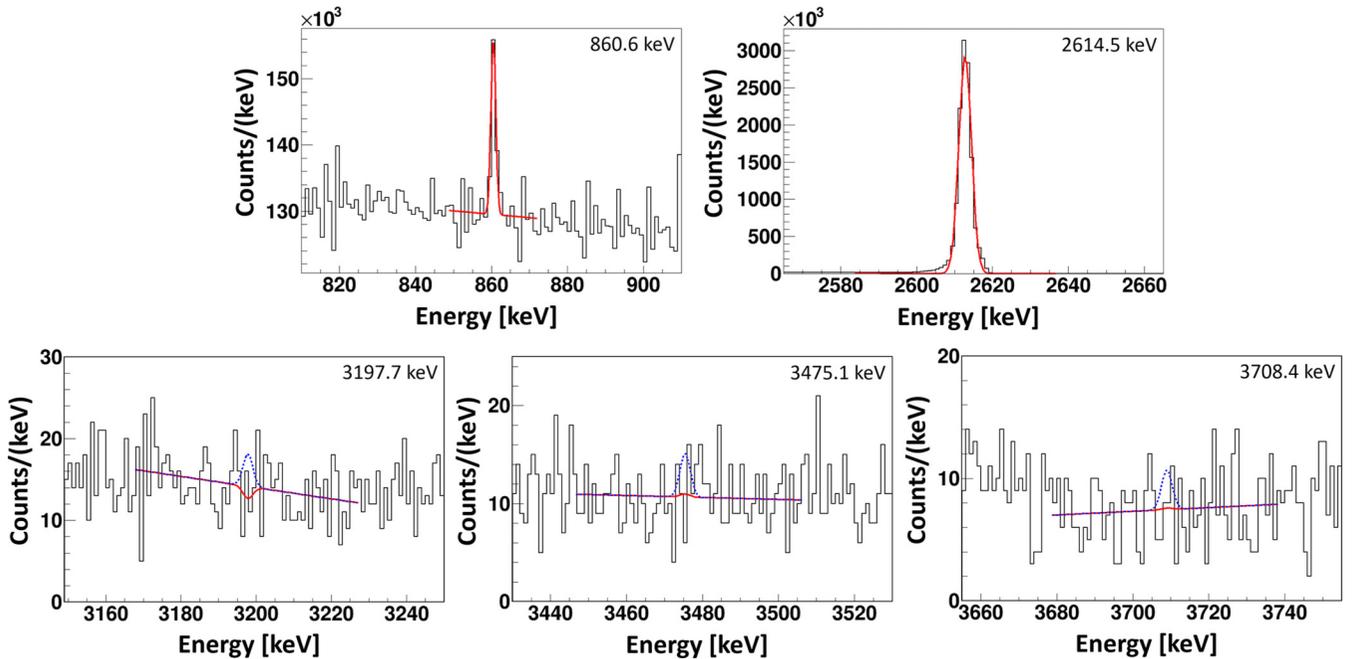


FIG. 8. The energy spectra for 39.5 days with fitting results at five energy regions, 860.6, 2614.5, 3197.7, 3475.1, and 3708.4 keV. The red lines are the best-fit curves, and the blue dotted lines show the limit estimation curves with 90% C.L.

the three peaks of interest, zero were rejected by the pileup cut.

The spectrum was calibrated for energy using a ^{60}Co source along with the data from the 0.1 g ThO_2 source. The energy resolution was calibrated using a multi-gamma source and a standard three-parameter resolution model $[(\sigma/E)^2 = (p_1/E)^2 + (p_2)^2 + (p_3/\sqrt{E})^2]$ and with fit values and errors extrapolated to the energies of interest. Based on agreement between calibration and data, peak position and Gaussian width errors were conservatively assigned as 0.6 keV and 0.5 keV respectively for the three peaks of interest. Fitting was performed with the centroid and width parameters floated and constrained by corresponding Gaussian constraint terms.

Figure 8 shows fitting results for five peaks at 860.6, 2614.5, 3197.7, 3475.1, and 3708.4 keV. In this measurement, only 860.6 and 2614.5 keV peaks were observed. Count rates for the 860.6 and 2614.5 keV peaks were $1.24(2) \times 10^{-2}$ and $3.56(0) \text{ s}^{-1}$, respectively. Upper limits on count rates for the 3197.7, 3475.1, and 3708.4 keV peaks were 3.84×10^{-6} , 4.53×10^{-6} , and $3.38 \times 10^{-6} \text{ s}^{-1}$, respectively (90% C.L.). In order to convert the count rates into activities, it is necessary

to know the absolute detection efficiency, which depends on γ energy. The detection efficiency can be estimated in two ways: Monte Carlo simulation and measurement using γ sources. Efficiencies were measured using rates of observed emission lines from the powder sample, with activities determined from the previously described measurement of the unshielded 0.1 g powder sample. For unshielded samples such as the 0.1 g powder, efficiency errors have been measured to be less than 10%. Efficiencies for the 3197.7, 3475.1 and 3708.4 keV peaks were determined using the GEANT4 simulation toolkit [23]. These could not be obtained from the measurement because of the unknown intensities of these transitions. Table I lists the simulated and measured efficiency values for several energies. Since we normalize the results of interest to the 2614.5 keV count rate, the relevant error is not the absolute efficiency error but the error in the ratios of the efficiencies at the energies of interest to the efficiency at 2614.5 keV for the shielded powder sample. We find that the Monte Carlo result agrees with the measured data within 11% at 2614.5 keV. Furthermore, agreement in measured and simulated ratios of efficiencies to the 2614.5 keV efficiency is better than 5%. The relevant γ cross sections change very slowly over the extrapolated energy region, but we conservatively assign a 10% systematic error to the efficiency ratios.

TABLE I. Detection efficiencies for the ThO_2 sample.

Energy (keV)	Simulation (%)	Measurement (%)
860.6	$1.80(18) \times 10^{-5}$	$2.10(92) \times 10^{-5}$
969.0	$1.89(19) \times 10^{-5}$	$2.29(23) \times 10^{-5}$
1621.0	$4.00(40) \times 10^{-4}$	$4.60(29) \times 10^{-4}$
2614.5	$6.76(68) \times 10^{-4}$	$7.52(48) \times 10^{-4}$
3197.7	$5.95(60) \times 10^{-4}$	
3475.1	$5.85(59) \times 10^{-4}$	
3708.4	$5.40(54) \times 10^{-4}$	

IV. RESULTS

Figure 9 shows an example of decay scheme for two of the excited states in the nucleus. In this decay scheme, the peak with energy E_3 is produced by two types of γ events: the single γ ray γ_3 and the true coincidence summing $E_1 + E_2$. Assuming γ_1 and γ_2 are emitted isotropically without angular correlations, C_3 , net counts of the E_3 energy peak, can be

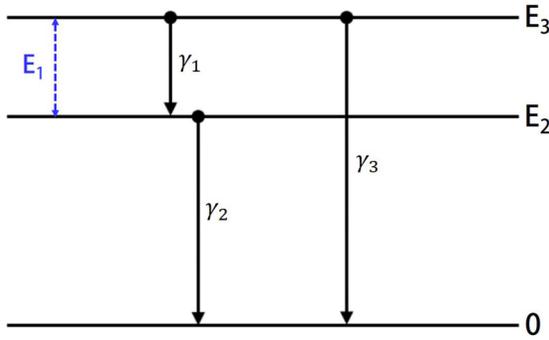


FIG. 9. An example decay scheme for two types of transitions which give the same energy from the E_3 state: the single deexcitation γ ray γ_3 and the true coincidence summing $\gamma_1 + \gamma_2$.

expressed as

$$C_3 = N_{\text{nu}} \times (I_3 \times \varepsilon_3 + I_1 \times \varepsilon_1 \times I_2 \times \varepsilon_2), \quad (2)$$

where I_i is the transition intensity, ε_i indicates the detection efficiency for each γ transition, and N_{nu} is the total number of emitted γ rays from the mother nucleus [15]. In this experiment, N_{nu} can be replaced with $N_{208\text{Tl}}$, the total number of all gamma-emitting events from the powder sample. From Eq. (2), intensity of the single- γ transition I_3 can be obtained by

$$I_3 = \frac{1}{\varepsilon_3} \times \left(\frac{C_3}{N_{208\text{Tl}}} - I_1 \times \varepsilon_1 \times I_2 \times \varepsilon_2 \right). \quad (3)$$

$N_{208\text{Tl}}$ can be calculated using the 2614.5 keV peak, which is produced by the single- γ transition with $E_\gamma = 2614.5$ keV. The net counts of the 2614.5 keV peak, C_{2615} , can be expressed by the product of its efficiency (ε_{2615}), intensity (I_{2615}), and $N_{208\text{Tl}}$. Therefore, $N_{208\text{Tl}}$ can be presented as

$$N_{208\text{Tl}} = \frac{C_{2615}}{\varepsilon_{2615} \times I_{2615}}. \quad (4)$$

With this substitution, we can examine the efficiency factors in Eq. (3) and see that by designing the experiment with sufficient shielding (low efficiencies) the coincidence term, $I_1 \times \varepsilon_1 \times I_2 \times \varepsilon_2$, can be made arbitrarily small relative to the first term, or, more importantly, relative to the error in the first term. For the data presented here, the second term in Eq. (3) was at least 50 times smaller than the uncertainty in the first term and was neglected. After neglecting this term, Eq. (3), and Eq. (4), the single- γ transition intensity I_3 can be expressed as

$$I_3 = \frac{C_3 \times \varepsilon_{2615} \times I_{2615}}{\varepsilon_3 \times C_{2615}}. \quad (5)$$

The intensities for the 3197.7, 3475.1, and 3708.4 keV single- γ transitions were calculated using Eq. (5) and net counts from the measurement. In this work, only upper limits were established because no peaks were observed at the ROIs. The upper limits for intensities of the 3197.7, 3475.1, and 3708.4 keV γ transitions were $1.2 \times 10^{-4}\%$, $1.4 \times 10^{-4}\%$, and $1.2 \times 10^{-4}\%$, respectively.

TABLE II. Gamma intensity. Upper-limit values are 90% C.L.

E_γ (keV)	This work (%)	Previous study (%) [15]	Theoretical value (%)
3197.7	$<1.2 \times 10^{-4}$	$<7 \times 10^{-3}$	6.43×10^{-7}
3475.1	$<1.4 \times 10^{-4}$	$<3 \times 10^{-3}$	6.52×10^{-10}
3708.4	$<1.2 \times 10^{-4}$	$<4 \times 10^{-3}$	1.25×10^{-10}

V. DISCUSSION

The Weisskopf formula is a classical and useful theoretical approximation for the transition rate of a de-excitation γ ray resulting from the transition of a single proton from one state to another [24,25]. The 583.2 and 3197.7 keV γ rays are emitted from the second excited level at 3197.7 keV in ^{208}Pb . The 583.2 keV γ transition is dominated by the $E2$ transition, and its Weisskopf transition rate λ_{583} is $6.04 \times 10^9 \text{ s}^{-1}$. For the 3197.7 keV γ transition, the dominant transition mode is $E5$, and the rate λ_{3198} calculated to be 45.7 s^{-1} . The γ intensity value and transition rate λ are in a proportional relationship so long as the γ rays in question are emitted from the same excited state [26]. The γ intensities for ^{208}Pb with energies lower than 3 MeV are already well known by many experiments. Using these known values and the proportionalities obtained from the Weisskopf transition rates, the intensities for unobserved γ transitions can be estimated theoretically. The intensity of the 3197.7 keV γ transition can be calculated using the following equation:

$$I_{3198} = \frac{I_{583} \times \lambda_{3198}}{\lambda_{583}} = 6.43 \times 10^{-7} (\%). \quad (6)$$

Similarly, intensities of the 3475.1 keV and 3709.4 keV γ transitions can be calculated as follows:

$$I_{3475} = \frac{I_{860} \times \lambda_{3475}}{\lambda_{860}} = 6.52 \times 10^{-10} (\%) \quad (7)$$

and

$$I_{3709} = \frac{I_{511} \times \lambda_{3709}}{\lambda_{511}} = 1.25 \times 10^{-10} (\%), \quad (8)$$

respectively.

Table II lists intensities of the 3197.7, 3475.1, and 3708.4 keV γ transitions which were obtained by three different studies: this work, the previous study by Vasil'ev *et al.* [15], and the theoretical estimation. As shown in the table, new upper limits for the intensities of three γ transitions were obtained from this study with limit values that are at least 19 times lower than the previous results. However, our upper limits are larger by several orders of magnitude than the theoretical estimates. The theoretical estimations using the Weisskopf formula may have large uncertainties because the formula does not consider all the structure information of a particular nucleus. It is known that experimental results often do not agree with Weisskopf estimations [27]. The numbers from experiments and theoretical calculations are known to be as much as 1000 times different in some cases [28,29]. However, the γ transition rates by Weisskopf formula for ^{208}Pb are expected to predict the real values reasonably well, because

^{208}Pb is one of the doubly magic nuclei that are relatively well studied by theoretical models and experiments. Although the rates expected from the theoretical prediction are much lower than the limits of this work, our results represent an important improvement to experimentally verified constraints on the rates which have a direct impact on $0\nu\beta\beta$ decay search experiments.

VI. CONCLUSION

We measured the intensities of γ transitions with $E_\gamma > 3$ MeV from the second, third, and fourth excited states to the ground state in ^{208}Pb , in the decay of the ^{208}Tl nucleus. A 2 kg sample of ThO_2 powder was measured for 39.5 days by using an HPGe detector. Improved upper limits for intensities of the 3197.7, 3475.1, and 3708.4 keV γ transitions were

$1.2 \times 10^{-4}\%$, $1.4 \times 10^{-4}\%$, and $1.2 \times 10^{-4}\%$, respectively. These results are at least 19 times lower than the previous measurements by Vasil'ev *et al.* [15]. The simulation study for AMoRE [16] shows that these three γ transitions with $E_\gamma > 3$ MeV in ^{208}Pb states will not contribute any background for $0\nu\beta\beta$ decay search experiments if using new upper-limit values from this study. However, our upper limits are still higher than theoretical estimates. Further studies with much improved experimental conditions are needed to achieve sensitivities corresponding to the expected theoretical values.

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