

Observation of two-neutrino double electron capture in ^{124}Xe with XENON1T

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Two-neutrino double electron capture ($2\nu\text{ECEC}$) is a second-order weak-interaction process with a predicted half-life that surpasses the age of the Universe by many orders of magnitude¹. Until now, indications of $2\nu\text{ECEC}$ decays have only been seen for two isotopes^{2–5}, ^{78}Kr and ^{130}Ba , and instruments with very low background levels are needed to detect them directly with high statistical significance^{6,7}. The $2\nu\text{ECEC}$ half-life is an important observable for nuclear structure models^{8–14} and its measurement represents a meaningful step in the search for neutrinoless double electron capture—the detection of which would establish the Majorana nature of the neutrino and would give access to the absolute neutrino mass^{15–17}. Here we report the direct observation of $2\nu\text{ECEC}$ in ^{124}Xe with the XENON1T dark-matter detector. The significance of the signal is 4.4 standard deviations and the corresponding half-life of 1.8×10^{22} years (statistical uncertainty, 0.5×10^{22} years; systematic uncertainty, 0.1×10^{22} years) is the longest measured directly so far. This study demonstrates that the low background and large target mass of xenon-based dark-matter detectors make them well suited for measuring rare processes and highlights the broad physics reach of larger next-generation experiments^{18–20}.

The long half-life of double electron capture makes it extremely rare, and the process has escaped detection for decades. In $2\nu\text{ECEC}$, two protons in a nucleus are simultaneously converted into neutrons by the absorption of two electrons from one of the atomic shells and the emission of two electron neutrinos¹ (ν_e). After the capture of the two atomic electrons, mostly from the K shell²¹, the filling of vacancies results in a detectable cascade of X-rays and Auger electrons²². The nuclear binding energy Q released in the process (on the order of 1 MeV) is carried away mostly by the two neutrinos, which are not detected within the detector. Thus, the experimental signature appears in the kiloelectronvolt, rather than the megaelectronvolt, range. The process is illustrated in Fig. 1.

$2\nu\text{ECEC}$ is allowed in the standard model of particle physics and is related to double β decay as a second-order weak-interaction process. However, few experimental indications exist. Geochemical studies^{4,5} for ^{130}Ba and a direct measurement^{2,3} for ^{78}Kr quote half-lives of the order of 10^{20} – 10^{22} yr.

Even longer timescales are expected for a hypothetical double electron capture without neutrino emission ($0\nu\text{ECEC}$)^{16,17}. A detection of this decay would show that neutrinos are Majorana particles¹⁵ (that is, their own anti-particles) and could help us to understand the dominance of matter over antimatter in our Universe by means of leptogenesis²³. A Majorana nature would give access to the absolute neutrino mass, but only with theoretical nuclear-matrix-element calculations. A plethora of different nuclear models^{8–14} can also be applied to predict the $2\nu\text{ECEC}$ half-life; thus, its measurement would provide a vital experimental constraint for these models, as well as insight into double- β -decay processes on the proton-rich side of the nuclide chart.

Here we study the $2\nu\text{ECEC}$ decay of ^{124}Xe . Natural xenon is a radio-pure and scalable detector medium that contains about 1 kg of ^{124}Xe per tonne. ^{124}Xe undergoes $2\nu\text{ECEC}$ to ^{124}Te with²⁴ $Q = 2,857$ keV.

Because the amount of energy released by the recoiling nucleus is negligible (on the order of 10 eV) and the neutrinos carrying away the energy Q are undetected, only the X-rays and Auger electrons are measured. The total energy for double K-shell-electron capture²⁴ is 64.3 keV. This value has already been corrected for energy depositions that do not exceed the xenon excitation threshold²². Previous searches for the $2\nu\text{ECEC}$ decay of ^{124}Xe were carried out with gas proportional counters using enriched xenon⁶, as well as large detectors originally designed for dark-matter searches²⁵. The currently leading lower limit on the half-life of this decay comes from the XMASS collaboration at $T_{1/2}^{2\nu\text{ECEC}} > 2.1 \times 10^{22}$ yr (90% confidence level)⁷.

XENON1T²⁶ was built to detect interactions of dark matter in the form of weakly interacting massive particles (WIMPs) and has recently placed the most stringent limits on the coherent elastic scattering of WIMPs with xenon nuclei²⁷. XENON1T uses 3.2 t of ultra-pure liquid xenon (LXe), of which 2 t are within the sensitive volume of the time-projection chamber (TPC): a cylinder with diameter and height of about 96 cm and with walls of highly reflective polytetrafluoroethylene, equipped with 248 photomultiplier tubes (PMTs). The TPC is used for the measurement of the scintillation (S1) and ionization signals (S2) induced by particle interactions—the latter by converting ionization electrons into light by means of proportional scintillation. It provides calorimetry and three-dimensional position reconstruction and measures the scatter multiplicity.

The detector is shielded by the overburden due to its underground location at Laboratori Nazionali del Gran Sasso, by an active water Cherenkov muon veto²⁸ and by the LXe itself. All detector materials were selected to have low amounts of radioactive impurities and low radon emanation rates²⁹. In addition, the anthropogenic β -emitter ^{85}Kr was removed from the xenon inventory by cryogenic distillation³⁰. The combination of material selection, active background reduction and selection of an inner low-background fiducial volume in the data analysis results in an extremely low event rate of about 80 events $\text{keV}^{-1} \text{t}^{-1} \text{yr}^{-1}$. This makes XENON1T the most sensitive detector for $2\nu\text{ECEC}$ searches in ^{124}Xe at present.

The data presented here were recorded between 2 February 2017 and 8 February 2018 as part of a dark-matter search. Details on the detector conditions and signal corrections can be found in the original publication²⁷. The data quality criteria from the dark-matter analysis were applied, with the exception of those exhibiting low acceptance in the energy region of interest, around 60 keV. During the analysis, the data were blinded (that is, inaccessible for analysis) from 56 keV to 72 keV and unblinded only after the data quality criteria, fiducial volume and background model had been fixed. Datasets acquired after detector calibrations with an external $^{241}\text{AmBe}$ neutron source or a deuterium–deuterium-fusion neutron generator were removed to reduce the impact of radioactive ^{125}I , which was produced by the activation of ^{124}Xe during neutron calibrations and was taken out within a few days using the purification system. A pre-unblinding quantification of this removal using short-term calibration data led to a first reduction of the dataset to a live time of 214.3 d. This dataset was used to construct the background model. After unblinding, the long-term behaviour of ^{125}I

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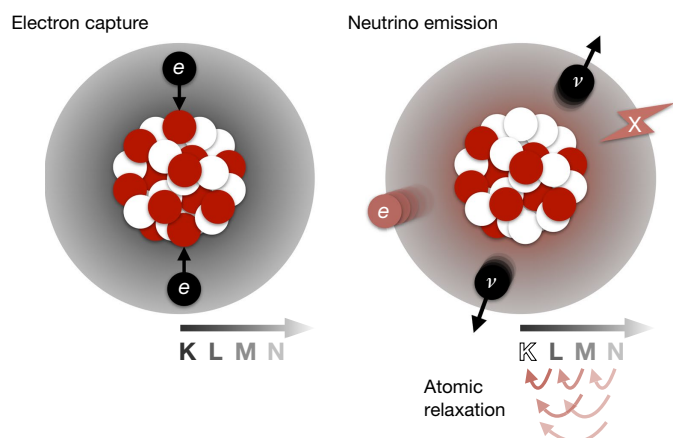


Fig. 1 | Schematic of two-neutrino double electron capture. In the $2\nu\text{ECEC}$ process the nucleus captures two atomic-shell electrons (black), usually from the K shell, and simultaneously converts two protons (red) to neutrons (white). Two neutrinos (black) are emitted in the nuclear process and carry away most of the decay energy while the atomic shell is left in an excited state with two holes in the K shell. A cascade of X-rays (red, 'X') and Auger electrons (red, 'e') are emitted during atomic relaxation, when the K shell is refilled from the higher-energy L, M and N shells. In turn, vacancies are created in the refilling shells and are refilled with electrons from the higher-energy shells (arrows).

could be quantified and led to a further removal of datasets (Methods). This yielded a final live time of 177.7 d.

Atomic X-rays and Auger electrons cannot be resolved individually owing to their sub-millimetre range in LXe and the rapid succession of the relevant atomic processes. Therefore, the experimental signature of K-shell $2\nu\text{ECEC}$ in XENON1T is a single S1 + S2 pair. Both S1 and S2 signals were used for the analysis to achieve the optimal energy resolution for the resulting peak. The energy scale around the expected signal at $E_0 = (64.3 \pm 0.6)$ keV was calibrated using monoenergetic lines of injected calibration sources (for example, $^{83\text{m}}\text{Kr}$), neutron-activated xenon isotopes and γ -rays from radioactive decays in the detector materials. The energy resolution of a Gaussian peak at E_0 is $\sigma/\mu = (4.1 \pm 0.4)\%$, where μ is the energy and σ is the width of the peak (Methods). The uncertainty on E_0 reflects the uncertainties of both the energy reconstruction and the correction for sub-excitation quanta. An ellipsoidal 1.5-t inner fiducial mass was identified as providing the optimal signal-to-background ratio in sideband studies between 80 keV and 140 keV, above the blinded signal region.

Understanding the measured energy spectrum is essential when searching for a small peak from $2\nu\text{ECEC}$. Three classes of backgrounds contribute to the spectrum: from intrinsic radioactive isotopes that are mixed with the LXe, from radioactive isotopes in the detector materials and from solar neutrinos. The latter is subdominant and well constrained from solar and nuclear physics. γ -rays from ^{60}Co and ^{40}K , as well as from ^{238}U and ^{232}Th decay chains, constitute the bulk of the detector material backgrounds. They can undergo forward Compton scattering before entering the 2.0-t active mass and produce a flat spectrum at low energies. Multiple scatters inside the active volume are rejected by selecting events with only a single S2 compatible with a single S1. The most important intrinsic background components are β decays of ^{214}Pb , a daughter of ^{222}Rn that is emanated from inner surfaces in contact with xenon, the two-neutrino double β decay of ^{136}Xe and the β decay of ^{85}Kr . Monoenergetic peaks from $^{83\text{m}}\text{Kr}$ injected for calibration and activation peaks that occur after neutron calibrations ($^{131\text{m}}\text{Xe}$ and $^{129\text{m}}\text{Xe}$) are present in the spectrum as well. The activation $^{124}\text{Xe} + n \rightarrow ^{125}\text{Xe} + \gamma$ has implications for $2\nu\text{ECEC}$ search, as ^{125}Xe decays to ^{125}I via electron capture. With a branching ratio of 100% and a half-life of 59.4 d, ^{125}I decays into an excited state of ^{125}Te . The subsequently emitted γ -ray together with the K-shell X-ray, which is produced in 87.5% of cases, leads to a monoenergetic peak at 67.3 keV. Owing to its proximity to E_0 , this peak would present a large background

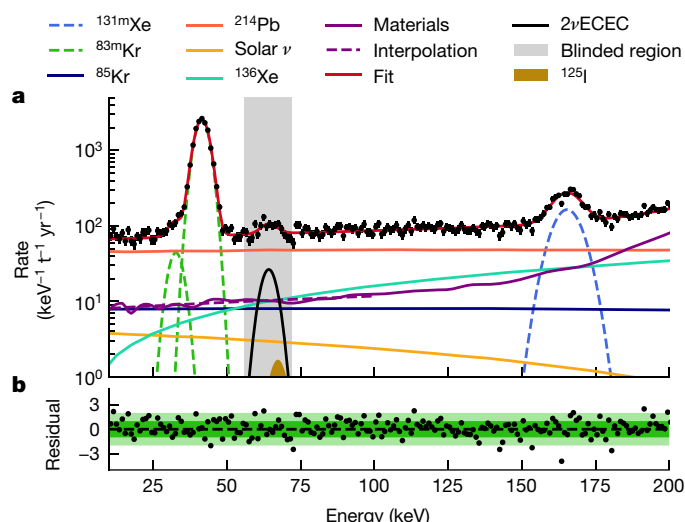


Fig. 2 | Fit of the background model to the measured energy spectrum.

The exposure of the background data is 177.7 d in the 1.5-t inner fiducial mass and the uncertainties are Poissonian. **a**, The data are described by a simultaneous fit of Monte Carlo-generated background spectra, taking into account all known background sources and the $2\nu\text{ECEC}$ signal (solid red line; $\chi^2/\text{d.o.f.} \approx 527.3/462$; d.o.f., degrees of freedom) in two sub-volumes of the detector (Methods). The linear interpolation of the detector material backgrounds below 100 keV is indicated by the purple dashed line. The remaining intrinsic background sources are shown as solid and dashed coloured lines. The energy region around the expected $2\nu\text{ECEC}$ peak was blinded (grey band) until the background model was defined. **b**, The residuals between the data and the fit, including the 1σ (2σ) bands, are shown in green (light green).

for the $2\nu\text{ECEC}$ search that would only become apparent after unblinding. Using an activation model based on the parent isotope, we verified that ^{125}I was removed from the detector with a time constant of $\tau = (9.1 \pm 2.6)$ d (Methods). This is in accordance with continuous xenon purification using hot zirconium getters²⁶. Accounting for artificial neutron activation from calibrations and for activation by radioactive thermal neutrons in the purification loop outside the water tank, we expect $N_{^{125}\text{I}} = (10 \pm 7)$ events in the 177.7-d dataset.

The background model was constructed by matching Monte Carlo simulations of all known background components¹⁸ with the measured energy spectrum. Taking into account the finite detector resolution, events with single energy depositions in the active volume were selected from the Monte Carlo data and convolved with the measured energy resolution. The weighted sum of all spectra was optimized simultaneously to resemble the measured energy spectrum (Methods). The blinded signal region was not used in the fit. The measured energy spectrum with the best fits for the individual components is shown in Fig. 2. After unblinding of the signal region, a clear peak at E_0 was identified. The energy and signal width obtained from the spectral fit to the unblinded data are $\mu = (64.2 \pm 0.5)$ keV and $\sigma = (2.6 \pm 0.3)$ keV, respectively. The resulting sum spectrum of the event rate is shown in Fig. 3. Converting the fit to the total event count yields $N_{^{125}\text{I}} = (9 \pm 7)$ events from the decay of ^{125}I and $N_{2\nu\text{ECEC}} = (126 \pm 29)$ events from $2\nu\text{ECEC}$. Compared to the null hypothesis, the $\sqrt{\Delta\chi^2}$ value of the best fit is 4.4.

Several consistency checks were carried out. We verified that the signal was homogeneously distributed in space and accumulated linearly with the exposure. A simultaneous fit of an inner (1.0 t) and an outer (0.5 t) detector mass with different background compositions yielded consistent signal rates. We verified the linearity of the energy calibration by identifying the ^{125}I activation peak at its expected position, which is separated from E_0 by more than the energy resolution.

The fit accounts for systematic uncertainties, such as cut acceptance and the number of ^{125}I events, by including them as fit-parameter

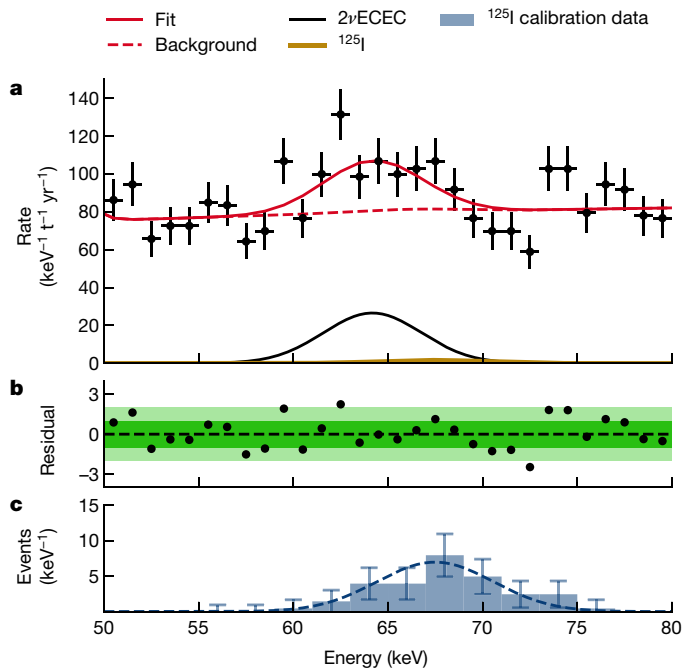


Fig. 3 | Zoom on the energy region of interest for $2\nu\text{ECEC}$ in ^{124}Xe . **a**, The best-fit contribution from $2\nu\text{ECEC}$ with $N_{2\nu\text{ECEC}} = 126$ events is shown by the solid black line and the full fit is indicated by the solid red line. The peak from ^{125}I with $N_{125\text{I}} = 9$ events is indicated by the solid gold line. The background-only model without $2\nu\text{ECEC}$ (red dashed line), again over the full-fit range, clearly does not describe the data. The error bars indicate Poisson uncertainties. **b**, Residuals for the best fit are given with the 1σ (2σ) band indicated in green (light green). **c**, A histogram of the ^{125}I activation peak as seen in 6 d of data after a dedicated neutron-generator calibration. The Poisson uncertainties (error bars) of the data were calculated before a linear background was subtracted. The peak shows the expected shift with respect to the $2\nu\text{ECEC}$ signal. The blue dashed line indicates a binned likelihood fit of the Gaussian peak.

constraints. Additional systematic uncertainties have to be considered when converting the observed number $N_{2\nu\text{ECEC}}$ into a half-life (Extended Data Table 1). The ^{124}Xe isotopic abundance in XENON1T was measured underground with a residual gas analyser with a systematic uncertainty of 1.5%. The resulting molar isotopic abundance of ^{124}Xe is $\eta = (9.94 \pm 0.14_{\text{stat}} + 0.15_{\text{sys}}) \times 10^{-4}$, which is 4% larger than the natural abundance³¹ of $\eta = (9.52 \pm 0.03) \times 10^{-4}$. The acceptance of the data selection criteria between 55 keV and 75 keV is constant within the uncertainties at $\epsilon = 0.967 \pm 0.007_{\text{stat}} \pm 0.033_{\text{sys}}$. The additional systematic uncertainty accounts for the fact that for a few data selection criteria only a lower limit on the acceptance was measurable. The finite resolution of the position reconstruction in XENON1T leads to an uncertainty on the fiducial mass. This was quantified by contrasting the mass fraction, derived from the fiducial volume geometry and LXe density³² of 2.862 g cm^{-3} at -96.1°C , with the fraction of $^{83\text{m}}\text{Kr}$ events in the fiducial volume. With this, the fiducial mass is $m = (1,502 \pm 9_{\text{sys}}) \text{ kg}$. The half-life is then calculated as

$$T_{1/2}^{2\nu\text{ECEC}} = \ln 2 \frac{\epsilon \eta N_A m t}{M_{\text{Xe}} N_{2\nu\text{ECEC}}}$$

where M_{Xe} is the mean molar mass of xenon, N_A is Avogadro's constant and t is the live time of the measurement. The resulting half-life for the K-shell double electron capture of ^{124}Xe is $T_{1/2}^{2\nu\text{ECEC}} = (1.8 \pm 0.5_{\text{stat}} \pm 0.1_{\text{sys}}) \times 10^{22} \text{ yr}$. This is the longest half-life ever measured directly. Indications of a similarly long half-life for $2\nu\text{ECEC}$ decay have been reported³ for ^{78}Kr . Within the uncertainties these half-lives are equally long, but the uncertainty of our result for ^{124}Xe is about two times smaller. Furthermore, our result is compatible

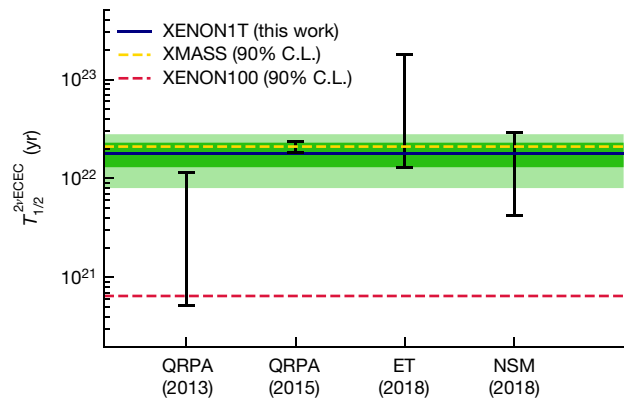


Fig. 4 | Observed $2\nu\text{ECEC}$ half-life compared with theoretical predictions and other experiments. The measured half-life (solid blue) with the 1σ (2σ) statistical uncertainty band indicated in green (light green) is compared to the experimental 90% confidence level (C.L.) upper limits from XMASS⁷ (dashed yellow line) and XENON100²⁵ (dashed red line). Recent results from nuclear structure calculations^{8,13,14} for the $2\nu\text{ECEC}$ show good agreement with the half-life measured in this work. The theoretical half-lives from the quasiparticle random-phase approximation (QRPA(2013)⁸ and QRPA(2015)¹³) were calculated for the double K-shell-electron (KK) capture (observed) as well as for electron captures from higher shells (not observed), so they have been scaled up by the KK fraction²¹ $f_{2\nu\text{KK}} = 0.767$ of the decay. The effective theory (ET¹⁴) and nuclear shell model (NSM¹⁴) half-lives for the double K-shell-electron capture are also shown.

with the lower limit from XMASS⁷ within the uncertainties (Fig. 4). With regard to nuclear theory, this measurement provides the first benchmark for nuclear structure models from the proton-rich side of the mass parabola. Predicted half-lives from recent nuclear calculations^{8,13,14}, which can now be refined further, are in the same window as the one observed (Fig. 4).

This first direct observation of $2\nu\text{ECEC}$ in ^{124}Xe also illustrates how xenon-based dark-matter search experiments, with their ever-growing target masses and simultaneously decreasing background levels, are becoming relevant for other rare event searches and neutrino physics. It sets the stage for $0\nu\text{ECEC}$ searches that can complement double- β -decay experiments in the hunt for the Majorana neutrino. Related processes involving the emission of one or two positrons ($2\nu\text{EC}\beta^+$, $2\nu\beta^+\beta^+$, $0\nu\text{EC}\beta^+$, $0\nu\beta^+\beta^+$) in ^{124}Xe might also exhibit interesting experimental signatures. The next-generation detectors XENONnT¹⁸, LZ¹⁹ and PandaX-4T³³ are already under construction and will be able to probe these as-yet-unobserved decays with unprecedented sensitivity.

Online content

Any methods, additional references, Nature Research reporting summaries, source data, statements of data availability and associated accession codes are available at <https://doi.org/10.1038/s41586-019-1124-4>.

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Competing interests The authors declare no competing interests.

Additional information

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XENON Collaboration*

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METHODS

Selection of the fiducial mass. Because the 2ν ECEC signal is proportional to the number of ^{124}Xe nuclei, it grows linearly with the xenon mass of the volume selected for the analysis, m_{volume} . The ability to distinguish signal events from background depends on the background uncertainty $\Delta N_{\text{background}}$. For a counting experiment, the uncertainty on the number of background events $N_{\text{background}}$ is of Poissonian nature, so $\Delta N_{\text{background}} = \sqrt{N_{\text{background}}}$. The discovery sensitivity in a detector volume S_{vol} is then proportional to the xenon mass in the selected volume divided by the background uncertainty:

$$S_{\text{vol}} \propto \frac{m_{\text{volume}}}{\sqrt{N_{\text{background}}}} \quad (1)$$

The S_{vol} parameter was optimized using an automated algorithm that tests both cylindrical and superellipsoidal volumes. A 1,502-kg-mass superellipsoid was found to give the optimal sensitivity. Because the signal region was blinded, the optimization was carried out in an energy sideband from 80 keV to 140 keV. For the fit of Monte Carlo simulations to the measured energy spectrum and consistency checks, the volume was segmented into an inner and outer volume (as indicated in Extended Data Fig. 1). Intrinsic background sources mixed with the xenon, solar neutrinos and the 2ν ECEC signal are expected to show the same activity in both volumes. However, the contribution from detector material backgrounds is strongest near the outer surfaces of these volumes. Fitting both volumes simultaneously gives a more robust fit and higher sensitivity than fitting a single monolithic volume. **Energy calibration and resolution.** Monoenergetic lines from the γ decays of four different isotopes were used for the energy calibration of the XENON1T detector. $^{83\text{m}}\text{Kr}$ is a gaseous calibration source that is homogeneously distributed inside the detector³⁴. The isomer undergoes a multi-step decay that is highly converted and deposits 41.5 keV inside the detector. This represents the lowest monoenergetic calibration point. The metastable isotopes $^{131\text{m}}\text{Xe}$ (163.9 keV) and $^{129\text{m}}\text{Xe}$ (236.2 keV) were neutron-activated during the calibration campaigns and decay with half-lives of 11.86 d and 8.88 d, respectively. The 1,173.2-keV and 1,332.5-keV transitions of ^{60}Co , which is present in the stainless steel detector components, such as the cryostat, are the highest energy calibration lines. Only energy depositions where the total energy of the γ transition is deposited in a single resolvable interaction within the detector—that is, the full absorption peak—were taken into account. The S1 and S2 signals from these interactions were then used to determine the yields of light and charge per unit energy for each source. These two quantities are anti-correlated^{35,36}, resulting in:

$$E = W \times \left(\frac{cS1}{g_1} + \frac{cS2_b}{g_2} \right) \quad (2)$$

at a given energy E . Here, $W = (13.7 \pm 0.2)$ eV is the average energy³⁷ needed to generate measurable quanta in LXe (S1 photons or S2 electrons), and $cS1$ and $cS2_b$ are the measured S1 and S2 signals corrected for detector effects. S1 is corrected for the spatially dependent S1 light collection efficiency, whereas S2 is corrected for the spatial dependencies of both the charge amplification and the S2 light collection efficiency. The subscript on $cS2_b$ identifies the S2 signal seen by the bottom PMT array, which was used for energy reconstruction to minimize the impact of signal saturation and non-uniformity due to single inactive PMTs in the top array. A fit to the measured data points gives the detector-specific calibration parameters g_1 and g_2 . The calibration procedure was carried out in ten slices along the central axis of the cylindrical detector to account for the depth dependence of $g_1(z)$ and $g_2(z)$ for the energy reconstruction.

The energy resolution was determined from the reconstructed spectrum by fitting Gaussian functions with the mean energy μ_E and standard deviation σ_E to monoenergetic peaks of the calibration sources ($^{83\text{m}}\text{Kr}$, $^{131\text{m}}\text{Xe}$, $^{129\text{m}}\text{Xe}$) and radioactive isotopes in the TPC materials (^{214}Pb , ^{208}Tl) up to 510.8 keV. The relative resolution is then given by σ_E/μ_E for each peak. Finally, the data points are fitted with a phenomenological function:

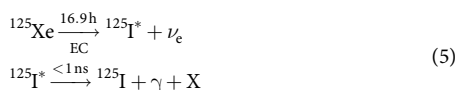
$$\frac{\sigma_E}{\mu_E} = \frac{a}{\sqrt{E}} + b \quad (3)$$

which gives an energy resolution of 4.1% at the 2ν ECEC energy (Extended Data Fig. 2).

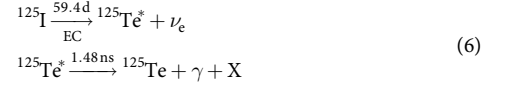
Iodine removal. Thermal neutrons can be captured by ^{124}Xe , producing ^{125}Xe :



^{125}Xe decays to ^{125}I via electron capture with a half-life of 16.9 h:



where X denotes the X-rays and Auger electrons emitted from the atomic relaxation after the electron capture. Iodine also undergoes electron capture to ^{125}Te with a 59.4-d half-life:



Both decays populate short-lived excited nuclear states of ^{125}I and ^{125}Te , and the signals from the γ transitions are merged with the atomic relaxation signals following the electron capture. The Te K-shell X-ray, which has a branching ratio of 87.5%, is merged with a 35.5-keV nuclear transition. This is problematic because it creates a Gaussian line centred around 67.3 keV, which is about 1σ away from the 64.3 keV expected for 2ν ECEC.

Two main mechanisms leading to the presence of ^{125}I in the detector have been identified: (1) artificial activation during calibration campaigns using neutrons from the deuterium–deuterium–fusion neutron generator or the $^{241}\text{AmBe}$ source and (2) activation outside the water shield by environmental thermal neutrons. As the decay rate of ^{125}Xe can be monitored during and after calibration, one can predict the decay rate of its iodine daughter. For environmental neutrons, flux measurements at Laboratori Nazionali del Gran Sasso were used to estimate the activation. These estimates were cross-checked with the ^{125}Xe decay peaks in the data. In the analysis of both the AmBe and the neutron generator data, fewer iodine decays than expected from the decay of the mother isotope ^{125}Xe were found. This is attributed to the removal of ^{125}I during the continuous purification of the detector's xenon inventory by circulation over hot zirconium getters. Owing to the blinding of the signal region containing the ^{125}I peak, the long-term behaviour of this removal could only be assessed after unblinding.

Because every ^{125}Xe decay in the detector leads to the presence of an ^{125}I nucleus, a model was constructed for the expected iodine decay rate from artificial activation by integrating the background-subtracted ^{125}Xe rate over time in one-day steps. The data were then convolved with the effective decay constant τ and fitted with a free amplitude and a linear background to the measured ^{125}I rate evolution in a 2σ interval around the peak (61.7–72.9 keV). An effective ^{125}I decay constant of $\tau = (9.1 \pm 2.6)$ d was found, which is in agreement with the expected decay constant from completely efficient getter removal.

Because the model was constructed directly from data, the uncertainties from the ^{125}Xe rates were propagated by introducing artificial Poisson fluctuations to the data points. An ^{125}I model was made for each variation of the ^{125}Xe data and fitted to the ^{125}I rate evolution. The best fit to the ^{125}I rate over time in 10-d bins and the uncertainty band derived from an ensemble of 1,000 fits are shown in Extended Data Fig. 3. Different binnings of 1–14 d were tested for consistency with χ^2 and log-likelihood fits.

Integration of each model over the actual data-taking periods yielded an expected number of ^{125}I decays $N_{125\text{I,art}}$. The ensemble distribution of $N_{125\text{I,art}}$ allowed us to extract both a central value and uncertainties. Only datasets with decay rates at the non-activated background level were selected for the 2ν ECEC search. The final data selection is shown in Extended Data Fig. 3. For the spectral fit of the remaining live time of 177.7 d we constrained the number of expected iodine events from artificial activation $N_{125\text{I,art}}$ using the model. We also constrained the radiogenic component $N_{125\text{I,rad}}$ by taking into account the effective decay constant τ .

Fitting method and result. The data were fitted with all known background sources, either simulated or modelled as Gaussian peaks, and the 2ν ECEC peak. The scaling parameters of the simulated Monte Carlo spectra and the properties of the Gaussian peaks are the fit parameters in the χ^2 minimization:

$$\chi^2_{\text{combined}}(\mathbf{p}) = \sum_i \frac{[R_i - f(E_i, \mathbf{p})]^2}{(\Delta R_i)^2} \quad (7)$$

where R_i is the measured event rate in the energy bin E_i and $f(E_i, \mathbf{p})$ is the background fit function. At energies below 100 keV, the low statistics of the simulated backgrounds from the detector construction materials require an interpolation of the simulated spectra in order to avoid over-fitting. Because the main background contribution from materials in this energy region is from single Compton scatters from γ -rays in the sensitive volume, a featureless spectrum is expected. Therefore, the sum of material contributions was linearly interpolated up to 100 keV. This gives:

$$\begin{aligned} f(E_i, \mathbf{p}) = & \left[\sum_k^{\text{materials}} p_k R_k(E_i) \right]_{\text{interpolated} < 100\text{ keV}} + \sum_l^{\text{intrinsic}} p_l R_l(E_i) \\ & + \sum_m^{\text{Gaussians}} \text{Gaussian}_m(\mathbf{p}_m, E_i) \end{aligned} \quad (8)$$

where the sums correspond to the interpolated material component, the intrinsic sources plus solar neutrinos and the Gaussian peaks, with fit parameters $p_{k,l,m} \in \mathbf{p}$. Knowledge from external measurements, such as material screening²⁹, ⁸⁵Kr concentration measurements²⁷ and elemental abundances, were incorporated into the fit function and constrained using terms of the form:

$$\text{constraint}_j = \frac{(\text{parameter}_j - \text{expectation}_j)^2}{\text{uncertainty}_j^2} \quad (9)$$

A deviation of the fit parameter from the expectation by $n \times \sigma$ will thus increase the value of the χ^2 function by n^2 . The Gaussian signal peak was constrained in the fit as well given the prior information on the expected position and width. Moreover, systematic uncertainties from the cut acceptance and fiducial mass were addressed by including these as constrained fit parameters in the fit function. As the fit was carried out in an inner (1.0 t; fit range 10–300 keV) and outer (0.5 t; fit range 10–200 keV) detector volume, each of the two volumes has its own χ^2 function with distinct parameters for the respective fiducial masses V and cut acceptances κ . The energy reconstruction was found to agree within the uncertainties. The full χ^2 function can then be written as:

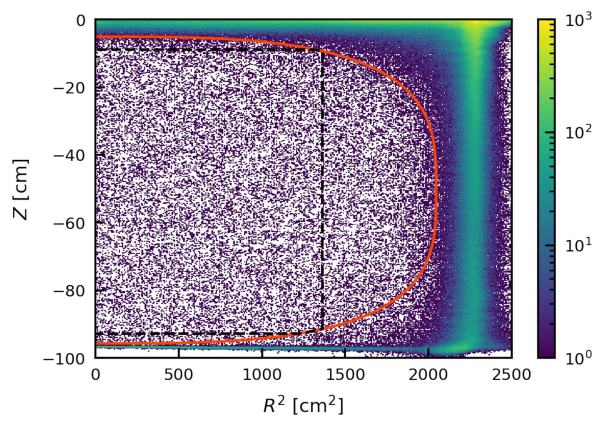
$$\chi_{\text{combined}}^2(\mathbf{p}, \mathbf{V}, \kappa) = \chi_{\text{inner}}^2(\mathbf{p}, V_{\text{inner}}, \kappa_{\text{inner}}) + \chi_{\text{outer}}^2(\mathbf{p}, V_{\text{outer}}, \kappa_{\text{outer}}) + \text{constraint}_p + \text{constraint}_V + \text{constraint}_\kappa \quad (10)$$

More details of the background modelling will be provided in a future publication. The χ^2 curve for the number of observed $2\nu\text{ECEC}$ events is shown in Extended Data Fig. 4. The 4.4σ significance is derived from the difference in $\Delta\chi^2$ between the best fit and a null result along the curve.

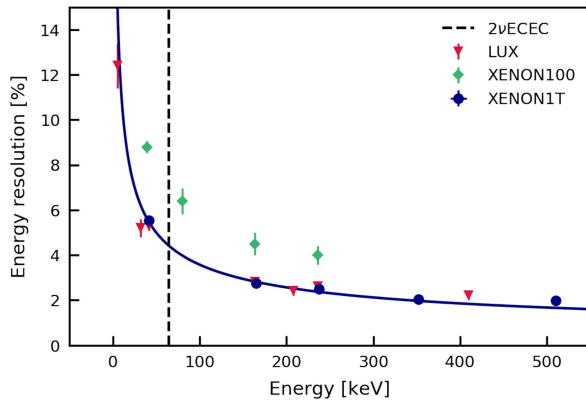
Data availability

The data that support the findings of this study are available from the corresponding authors upon reasonable request.

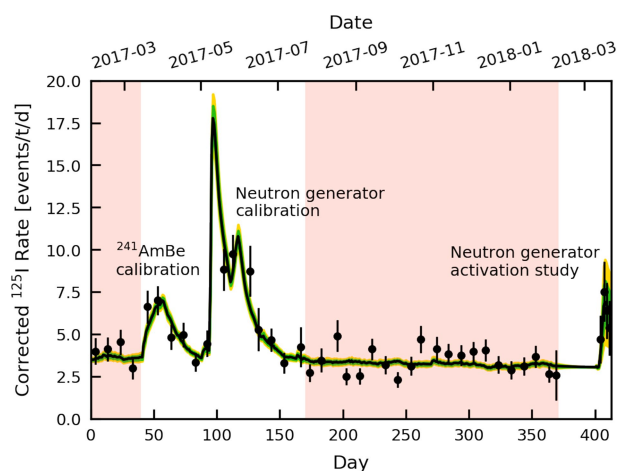
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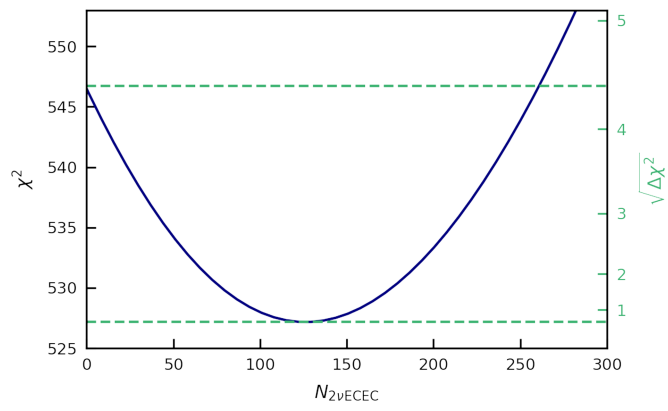
Extended Data Fig. 1 | Spatial distribution of events. Interaction depth Z versus squared radius R^2 for events with energies 80–140 keV. High-density areas correspond to the edges of the TPC, where most of external β and γ radiation is absorbed. The 1,502-kg fiducial volume is indicated by the solid red line. Further segmentation into an inner (1.0 t) and an outer (0.5 t) volume is marked by the black dashed line.



Extended Data Fig. 2 | Energy resolution. Ratio of the mean peak energy (μ_E) to the peak width (σ_E) for low-energy monoenergetic lines in selected LXe dark-matter experiments (LUX³⁸ and XENON100³⁹) and in the 1.5-t fiducial mass of the XENON1T detector. The relative resolution is defined as the σ_E/μ_E ratio of the Gaussian lines and is fitted using a phenomenological function (solid blue line). For XENON1T the data points are ^{83m}Kr (41.5 keV), ^{131m}Xe (163.9 keV), ^{129m}Xe (236.2 keV), ^{214}Pb (351.9 keV) and ^{208}Tl (510.8 keV). Only statistical uncertainties are shown for XENON1T (smaller than the markers). The energy of the $2\nu\text{ECEC}$ peak is indicated by the black dashed line.



Extended Data Fig. 3 | ^{125}I time evolution. Fit of the ^{125}I model to data in a 2σ energy interval around the mean energy of the ^{125}I peak in 10-d bins with Poisson uncertainties. Periods with an increased ^{125}I decay rate are attributed to artificial activations from neutron calibrations, equipment tests and a dedicated activation study. The decrease of the rate to the background level corresponds to an effective iodine decay constant of $\tau = 9.1$ d. The best fit is shown as a solid black line. The green (yellow) bands mark the 1σ (2σ) model uncertainties resulting from the Poisson uncertainties of the ^{125}Xe data underlying the model. The pink bands indicate the data selection for the $2\nu\text{ECEC}$ search, where the decay rate has returned to the background level.



Extended Data Fig. 4 | χ^2 curve for the number of measured 2ν ECEC events. By comparing the best-fit value of $N_{2\nu\text{ECEC}} = 126$ events to a null result one obtains $\sqrt{\Delta\chi^2} = 4.4$.

Extended Data Table 1 | Systematic uncertainties

a) Variable in $T_{1/2}^{\text{ECEC}}$ calculation	Uncertainty [%]	
Fiducial mass m	0.6	
ROI cut acceptance ϵ	3.4	
^{124}Xe abundance η	1.5	
b) Constrained fit parameter	Value $\pm$ uncertainty	Parameter pull [σ]
ν_{solar} multiplier	1.00 ± 0.20	0.3
^{136}Xe $2\nu\beta\beta$ multiplier	1.00 ± 0.05	−0.2
Volume _{inner,outer} multipliers	1.00 ± 0.01	$0.7_{\text{inner}}, -0.7_{\text{outer}}$
High energy acceptance _{inner,outer} multipliers	0.67 ± 0.33	$0.1_{\text{inner}}, -1.0_{\text{outer}}$
^{85}Kr concentration	(0.66 ± 0.12) ppt $^{\text{nat}}\text{Kr}/\text{Xe}$	0.3
$N_{125\text{I}}$	(10 ± 7) events	−0.2
$\mu_{125\text{I}}$	(67.3 ± 0.5) keV	−0.1
$\sigma_{125\text{I}}$	(2.8 ± 0.5) keV	−0.1
$\mu_{2\nu\text{ECEC}}$	(64.3 ± 0.6) keV	−0.3
$\sigma_{2\nu\text{ECEC}}$	(2.6 ± 0.3) keV	−0.2
$\mu_{83\text{mKr},1}$	(32.2 ± 0.6) keV	0.7
$\mu_{83\text{mKr},2}$	(41.5 ± 0.6) keV	−0.1
$\mu_{131\text{mXe}}$	(163.9 ± 0.6) keV	2.4
$\mu_{129\text{mXe}}$	(236.2 ± 0.6) keV	1.0

a. Uncertainties in the half-life calculation are given as percentages of the corresponding variable values. **b.** Systematic uncertainties incorporated as fit constraints are given in the unit used in the fit. All parameters are shared between the χ^2 functions for both volumes, with the exception of the volume and high-energy acceptance multipliers. The volume multipliers are chosen such that the fitted high-energy acceptance ranges between the lower limit derived from the data and unity. The parameter pulls of the fit are given in units of the uncertainty σ . ROI, region of interest; ppt, parts per trillion; $T_{1/2}^{\text{ECEC}}$, $2\nu\text{ECEC}$ half-life.