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

XENON Collaboration*

Two-neutrino double electron capture ($2\nu$ECEC) is a second-order weak-interaction process with a predicted half-life that surpasses the age of the Universe by many orders of magnitude. Until now, indications of $2\nu$ECEC decays have only been seen for two isotopes $^{78}$Kr and $^{130}$Ba, and instruments with very low background levels are needed to detect them directly with high statistical significance. The $2\nu$ECEC half-life is an important observable for nuclear structure models 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. Here we report the direct observation of $2\nu$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 $1.8 \times 10^{22}$ years (statistical uncertainty, $0.5 \times 10^{22}$ years; systematic uncertainty, $0.1 \times 10^{22}$ 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.

The long half-life of double electron capture makes it extremely rare, and the process has escaped detection for decades. In $2\nu$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 neutrinos ($\nu_e$). After the capture of the two atomic electrons, mostly from the K shell, the filling of vacancies results in a detectable cascade of X-rays and Auger electrons. The nuclear binding energy $Q$ released in the process (on the order of 1 MeV) is carried away mostly by the two neutrons, which are not detected within the detector. Thus, the experimental signature appears in the kielelectronvolt, rather than the megaelectronvolt, range. The process is illustrated in Fig. 1.

$2\nu$ECEC is allowed in the standard model of particle physics and is related to double $\beta$ decay as a second-order weak-interaction process. However, few experimental indications exist. Geochemical studies for $^{130}$Ba and a direct measurement 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 (0t/ECEC). A detection of this decay would show that neutrinos are Majorana particles and could help us to understand the dominance of matter over antimatter in our Universe by means of leptogenesis. 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 can also be applied to predict the $2\nu$ECEC half-life; thus, its measurement would provide a vital experimental constraint for these models, as well as insight into double-$\beta$-decay processes on the proton-rich side of the nuclide chart.

Here we study the $2\nu$ECEC decay of $^{124}$Xe. Natural xenon is a radiopure and scalable detector medium that contains about 1 kg of $^{124}$Xe per tonne. $^{124}$Xe undergoes $2\nu$ECEC to $^{124}$Te with $Q = 2.857$ 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 capture is 64.3 keV. This value has already been corrected for energy depositions that do not exceed the xenon excitation threshold. Previous searches for the $2\nu$ECEC decay of $^{124}$Xe were carried out with gas proportional counters using enriched xenon, as well as large detectors originally designed for dark-matter searches. The currently leading lower limit on the half-life of this decay comes from the XMASS collaboration at $T_{1/2}^{\nu ECEC} > 2.1 \times 10^{22}$ yr (90% confidence level).

XENON1T 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 nuclei. 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 veto and by the LXe itself. All detector materials were selected to have low amounts of radioactive impurities and low radon emission rates. In addition, the anthropogenic $^{85}$Kr was removed from the xenon inventory by cryogenic distillation. 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 keV$^{-1}$ t$^{-1}$ yr$^{-1}$. This makes XENON1T the most sensitive detector for $2\nu$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 publication. 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 $^{241}$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
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 Lx and the rapid succession of the relevant atomic processes. Therefore, the experimental signature of K-shell 2νECEC 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 $E_0 = (64.3 \pm 6.0)$ keV was calibrated using monoenergetic lines of injected calibration sources (for example, $^{83m}$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 $\mu$ is the energy and $\sigma$ 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 2νECEC. Three classes of backgrounds contribute to the spectrum: from intrinsic radioactive isotopes that are mixed with the Lx, from radioactive isotopes in the detector materials and from solar neutrinos. The latter is subordinate 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 $^{83m}$Kr injected for calibration and activation peaks that occur after neutron calibrations ($^{131m}$Xe and $^{129m}$Xe) are present in the spectrum as well. The activation $^{125}$Xe + n $\rightarrow$ $^{125}$Xe + γ has implications for 2νECEC 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 for the 2νECEC 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 radionuclides in the purification loop outside the water tank, we expect $N_{125I} = (10 \pm 7)$ 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 $N_{125I} = (9 \pm 7)$ events from the decay of $^{125}$I and $N_{2νECEC} = (126 \pm 29)$ events from 2νECEC. Compared to the null hypothesis, the $\sqrt{\chi^2}$ 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...
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 calculations8,13,14 for the 2νECEC 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 fraction13 (\(J_{KK}=0.767\)) 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 calculations8,13,14, which can now be refined further, are in the same window as the one observed (Fig. 4).

This first direct observation of 2νECEC 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 0νECEC 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 (2νEC), 2νββ, 0νEC, and 0νββ) 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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METHODS

Selection of the fiducial mass. Because the $2\nu$ECEC signal is proportional to the number of $^{125}$Xe nuclei, it grows linearly with the xenon mass of the volume selected for the analysis, $m_{\text{volume}}$. 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 $S_{\text{vol}}$ is then proportional to the xenon mass in the selected volume divided by the background uncertainty:

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

The $S_{\text{vol}}$ 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 neutrons and the $2\nu$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 $\gamma$ decays of four different isotopes were used for the energy calibration of the XENON1T detector. $^{85m}$Kr is a gaseous calibration source that is homogeneous distributed inside the detector. 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 $^{131m}$Xe (163.9 keV) and $^{128m}$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 $^{90}$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 $\gamma$ 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, resulting in:

$$E = W \times \left( \frac{cS1}{g1} + \frac{cS2}{g2} \right)$$  \hspace{1cm} (2)

at a given energy $E$. Here, $W = (13.7 \pm 0.2) \text{ eV}$ is the average energy needed to generate measurable quanta in LXe ($S1$ photons or $S2$ electrons), and $cS1$ and $cS2$ 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, whereas $S2$ 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 $cS2$ identifies the $S2$ signal seen by the bottom PMT.
where the sums correspond to the interpolated material component, the intrinsic sources plus solar neutrinos and the Gaussian peaks, with fit parameters \( p_{\text{fit}}, m \in p \).

Knowledge from external measurements, such as material screening\(^{29}\), \(^{83}\)Kr concentration measurements\(^{27}\) and elemental abundances, were incorporated into the fit function and constrained using terms of the form:

\[
\text{constraint}_j = \frac{(\text{parameter}_j - \text{expectation})^2}{\text{uncertainty}_j^2}
\]

(9)

A deviation of the fit parameter from the expectation by \( n \times \sigma \) will thus increase the value of the \( \chi^2 \) 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 \( \chi^2 \) function with distinct parameters for the respective fiducial masses \( V \) and cut acceptances \( \kappa \). The energy reconstruction was found to agree within the uncertainties. The full \( \chi^2 \) function can then be written as:

\[
\chi^2_{\text{combined}}(p, V, \kappa) = \chi^2_{\text{inner}}(p, V_{\text{inner}}, \kappa_{\text{inner}}) + \chi^2_{\text{outer}}(p, V_{\text{outer}}, \kappa_{\text{outer}}) + \text{constraint}_p + \text{constraint}_V + \text{constraint}_\kappa
\]

(10)

More details of the background modelling will be provided in a future publication.

The \( \chi^2 \) curve for the number of observed \( 2\nu \text{ECEC} \) events is shown in Extended Data Fig. 4. The 4.4\( \sigma \) 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 $\beta$ and $\gamma$ 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 ($\mu_E$) to the peak width ($\sigma_E$) for low-energy monoenergetic lines in selected LXe dark-matter experiments (LUX\textsuperscript{38} and XENON100\textsuperscript{39}) and in the 1.5-t fiducial mass of the XENON1T detector. The relative resolution is defined as the $\sigma_E/\mu_E$ 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$\nu$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 $2\sigma$ 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 $1\sigma$ ($2\sigma$) model uncertainties resulting from the Poisson uncertainties of the $^{125}$Xe data underlying the model. The pink bands indicate the data selection for the $2\nu$ECEC search, where the decay rate has returned to the background level.
Extended Data Fig. 4 | $\chi^2$ curve for the number of measured $2\nu$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 $T^{\text{ECEC}}_{1/2}$ calculation | Uncertainty [%] |
|-----------------------------------------------|-----------------|
| Fiducial mass $m$                             | 0.6             |
| ROI cut acceptance $\epsilon$                | 3.4             |
| $^{124}\text{Xe}$ abundance $\eta$           | 1.5             |

| b) Constrained fit parameter                  | Value ± uncertainty | Parameter pull [$\sigma$] |
|-----------------------------------------------|---------------------|--------------------------|
| $\nu_{\text{solar}}$ multiplier              | $1.00 \pm 0.20$     | 0.3                      |
| $^{136}\text{Xe}$ 2$\nu\beta\beta$ multiplier | $1.00 \pm 0.05$     | -0.2                     |
| Volume$_{\text{inner,outer}}$ multipliers    | $1.00 \pm 0.01$     | 0.7$_{\text{inner}}$, -0.7$_{\text{outer}}$ |
| High energy acceptance$_{\text{inner,outer}}$ multipliers | $0.67 \pm 0.33$     | 0.1$_{\text{inner}}$, -1.0$_{\text{outer}}$ |
| $^{85}\text{Kr}$ concentration               | $(0.66 \pm 0.12)$ ppt $^{\text{nat}}\text{Kr}/\text{Xe}$ | 0.3                      |
| $N_{125\text{I}}$                            | $(10 \pm 7)$ events | -0.2                     |
| $\mu_{125\text{I}}$                          | $(67.3 \pm 0.5)$ keV | -0.1                     |
| $\sigma_{125\text{I}}$                       | $(2.8 \pm 0.5)$ keV | -0.1                     |
| $\mu_{2\nu\text{ECEC}}$                     | $(64.3 \pm 0.6)$ keV | -0.3                     |
| $\sigma_{2\nu\text{ECEC}}$                  | $(2.6 \pm 0.3)$ keV | -0.2                     |
| $\mu_{83\text{mKr},1}$                      | $(32.2 \pm 0.6)$ keV | 0.7                      |
| $\mu_{83\text{mKr},2}$                      | $(41.5 \pm 0.6)$ keV | -0.1                     |
| $\mu_{131\text{mXe}}$                       | $(163.9 \pm 0.6)$ keV | 2.4                      |
| $\mu_{129\text{mXe}}$                       | $(236.2 \pm 0.6)$ 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 $\chi^2$ 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 $\sigma$. ROI, region of interest; ppt, parts per trillion; $T^{\text{ECEC}}_{1/2}$, $2\nu\beta\beta$ ECEC half-life.