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New experimental limits on the Pauli-forbidden transitions in ^{12}C nuclei obtained with 485 days Borexino data

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The Pauli exclusion principle (PEP) has been tested for nucleons (n, p) in ^{12}C with the Borexino detector. The approach consists of a search for γ, n, p, and $\beta^{\pm}$ emitted in a non-Paulian transition of $1P_{3/2}$-shell nucleons to the filled $1S_{1/2}$ shell in nuclei. Due to the extremely low background and the large mass (278 tons) of the Borexino detector, the following most stringent up-to-date experimental bounds on PEP violating transitions of nucleons have been established: $\tau({}^{12}\mathrm{C} \to {}^{12}\widetilde{\mathrm{C}} + \gamma) \geqslant 5.0 \times 10^{31}$ yr, $\tau({}^{12}\mathrm{C} \to {}^{11}\widetilde{\mathrm{B}} + p) \geqslant 8.9 \times 10^{29}$ yr, $\tau({}^{12}\mathrm{C} \to {}^{11}\widetilde{\mathrm{C}} + n) \geqslant 3.4 \times 10^{30}$ yr, $\tau({}^{12}\mathrm{C} \to {}^{12}\widetilde{\mathrm{N}} + e^- + \widetilde{\nu}_e) \geqslant 3.1 \times 10^{30}$ yr, and $\tau({}^{12}\mathrm{C} \to {}^{12}\widetilde{\mathrm{B}} + e^+ + \nu_e) \geqslant 2.1 \times 10^{30}$ yr, all at 90% C.L. The corresponding upper limits on the relative strengths for the searched non-Paulian electromagnetic, strong and weak transitions have been estimated as $\delta^2_\gamma \leqslant 2.2 \times 10^{-57}$, $\delta^2_N \leqslant 4.1 \times 10^{-60}$, and $\delta^2_\beta \leqslant 2.1 \times 10^{-35}$.

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I. INTRODUCTION

The exclusion principle was formulated by W. Pauli in [1] and in its original form postulated that "there can never be two or more equivalent electrons in an atom" [1]. In the case of Bohr atoms it meant that only one electron with definite spin orientation can occupy each of the allowed orbits. This statement was later formalized in the framework of quantum mechanics by saying that for two identical electrons the total wave function is antisymmetric under electron permutation. In relativistic quantum field theory (QFT), the Pauli exclusion principle (PEP) appears automatically for systems of identical fermions as a result of the anticommutativity of the fermion creation and annihilation operators.

Although the PEP is of fundamental importance, its physical cause is not yet understood. According to Okun "a non-conformist approach to the PEP could be traced to Dirac and Fermi" [2]. Both Dirac and Fermi discussed the implications of a small PEP violation on atomic transitions and on atomic properties [3,4].

Experimental searches for possible PEP violations started about 15 years later when the electron stability was tested. Pioneering experiments were performed by Reines and Sobel by searching for x rays emitted in the transition of an L-shell electron to the filled K shell in an atom [5], and by Logan and Ljubicic, who searched for γ quanta emitted in a PEP-forbidden transition of nucleons in nuclei [6].

From 1987 to 1991 theoretical models implicating PEP violation were constructed by Ignatiev and Kuzmin [7], Greenberg and Mohapatra [8–10], and Okun [11], but it was shown by Govorkov [12] that even a small PEP violation leads to negative probabilities for some processes. Moreover, in 1980 Amado and Primakoff pointed out that in the framework of quantum mechanics PEP-violating transitions [5,6] are forbidden even if PEP violation takes place [13].

At present, no acceptable theoretical formalism exists. In particular, it is not possible to account for PEP violation by means of a self-consistent and noncontradictory "small" parameter, as in the case of P- and CP-symmetry violation or L and B nonconservation. The results of experiments are presented as lifetime limits or as limits on the relative strength of the normal and Pauli-forbidden transitions. Critical studies of the possible violation of PEP have been done both theoretically and experimentally in Refs. [2,14,15]. More reviews and references can be found in Refs. [16,17].

There are two (or four, if we consider electrons and nucleons separately) types of experiments to look for PEP violation. The first one is based on the search for atoms or nuclei in a non-Paulian state; the second one is based on the search for the prompt radiation accompanying non-Paulian transitions of electrons or nucleons.

Experiments of the first type have been performed by Novikov and co-workers [18,19] and Nolte *et al.* [20], who looked for non-Paulian exotic atoms of ^{20}Ne and ^{36}Ar with three electrons on the K shell using mass spectroscopy on fluorine and chlorine samples. Similarly, the atoms of Be with four electrons in the $1s$ state that look like He atoms were searched for by Javorsek *et al.* [21]. The anomalous carbon atoms in boron samples were searched for by γ-activation analysis by Barabash *et al.* [22]. The PEP-forbidden nuclei of ^{5}Li with three protons in the $1S$ shell was searched for by Nolte *et al.* [23], using time-of-flight mass spectroscopy .

Goldhaber was the first to point out that the same experimental data that were used to set a limit on the lifetime of the electron can be used to test the validity of the PEP for atomic electrons [5]. From the experimental point of view, the searches for characteristic x rays from electron decay inside an atomic shell [24–35] are often indistinguishable from the PEP-violating transition, but according to Amado and Primakoff [13] these transition do not take place even if PEP is violated. This restriction is not valid for transitions accompanied by a change of the number of identical fermions (e.g., non-Paulian $\beta^\pm$ transitions) and can be evaded in composite models of electrons or models including extra dimensions [8,36].

A new method, realized by Ramberg and Snow, looked for anomalous x rays emitted by Cu atoms in a conductor [37]. The established upper limit on the probability for the "new" electron passing in the conductor to form a non-Paulian atom with three electrons in the K shell is 1.7×10^{-26}. An improvement of the sensitivity of the method is currently being planned by the VIP Collaboration [38]. Laser atomic and molecular spectroscopy were used to search for anomalous PEP-forbidden spectral lines of ^{4}He atoms [39] and molecules of O_2 [40,41] and CO_2 [42].

The violation of PEP in the nucleon system has been studied by searching for the non-Paulian transitions with γ emission [43,44] (Kamiokande, NEMO-II), p emission [35,45,46] (Elegant-V, DAMA/LIBRA), n emission [47], non-Paulian β^+ and β^- decays [44,48] (LSD, NEMO-II), and in nuclear (p, p) and (p, α) reactions on ^{12}C [49].

The strongest limits for non-Paulian transitions in ^{12}C with γ, p, n, α, and $\beta^\pm$ emissions were obtained with a prototype of the Borexino detector: the Counting Test Facility (CTF) [50]. In this paper we present the new results obtained with 485 days of Borexino data. The large Borexino mass (70 times larger than the CTF one) and its extremely low background level (200 times lower than in CTF at 2 MeV) enabled us to improve the lifetime limits for non-Paulian transitions in ^{12}C by three to four orders of magnitude with respect to CTF.

II. EXPERIMENTAL SETUP AND MEASUREMENTS

A. Brief description of Borexino

Borexino is a real-time detector for solar neutrino spectroscopy located at the Gran Sasso Underground Laboratory. Its main goal is to measure low-energy solar neutrinos via (ν,e) scattering in an ultrapure liquid scintillator. The extremely high radiopurity of the detector and its large mass allow one to simultaneously address other fundamental questions of particle physics and astrophysics.

The main features of the Borexino detector and its components have been thoroughly described in Refs. [51–54]. Borexino is a scintillator detector with an active mass of 278 tons of pseudocumene (PC, C_9H_{12}), doped with 1.5 g/L of PPO ($C_{15}H_{11}NO$). The scintillator is inside a thin nylon vessel (IV, inner vessel) and is surrounded by two concentric PC buffers (323 and 567 tons) doped with a small amount of light quencher (dymethilpthalate, DMP) to reduce their scintillation. The scintillator and buffers are contained in a stainless steel sphere (SSS) with a diameter of 13.7 m. The two PC buffers are separated by a second thin nylon membrane to prevent diffusion of radon coming from photomultipliers (PMTs), light concentrators, and SSS walls toward the scintillator. The SSS is enclosed in a 18.0-m-diameter, 16.9-m-high domed water tank (WT), containing 2100 tons of ultrapure water as an additional shield against external γ rays and neutrons. The scintillation light is detected by 2212 8″ PMTs uniformly distributed on the inner surface of the SSS. All the internal components of the detector were selected following stringent radiopurity criteria. The WT is equipped with 208 additional PMTs that act as a Cherenkov muon detector (outer detector) to identify the residual muons crossing the detector.

B. Detector calibration and energy and spatial resolutions

In Borexino charged particles are detected by their scintillation light-producing interactions with the liquid scintillator. The energy of an event is measured by using the total collected light from all PMTs. In a simple approach, the response of the detector is assumed to be linear with respect to the energy released in the scintillator. The coefficient linking the event energy and the total collected charge is called the light yield (or photoelectron yield). Deviations from linearity at low energy can be taken into account by the ionization deficit function $f(k_B, E)$, where k_B is the empirical Birks' constant [55].

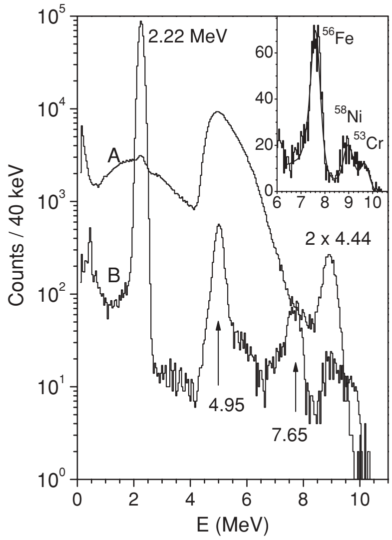

FIG. 1. The energy spectra of prompt (A) and delayed (B) signals registered with an ^{241}Am-^{9}Be source. In the inset, the γ lines from neutron captures on a stainless steel holder of an Am-Be source are shown.

The detector energy and spatial resolution were studied with radioactive sources placed at different positions inside the inner vessel. For relatively high energies (>2 MeV), which are of interest for non-Paulian transition studies, the energy calibration was performed with an ^{241}Am-^{9}Be neutron source. Figure 1 shows the spectrum obtained with the source placed at the center of the detector. The reactions $^{9}\mathrm{Be}(\alpha,n)^{12}\mathrm{C}_{\mathrm{gs}}$ and $^{9}\mathrm{Be}(\alpha,n)^{12}\mathrm{C}^{*}$ (4.44 MeV) produce two main neutron groups with energies up to 11 and 6.5 MeV, respectively. The resulting neutrons are thermalized by elastic and inelastic scattering in the hydrogen-rich organic scintillator and eventually are captured by protons or carbon nuclei. The upper (red) spectrum in Fig. 1 corresponds to the prompt neutrons and γ rays, whereas the lower (black) one is that of the delayed signals. The energy scale was determined with the 2.22- and 4.95-MeV γ de-excitations following neutron capture on ^{1}H and ^{12}C nuclei, and with the 8.88-MeV peak, sum of two 4.44-MeV γ quanta. The expected shift of the 8.88-MeV peak position (caused by the residual energy of the scattered neutron) is suppressed by the sizable quenching factor of low-energy protons. The 7.65-MeV γ line following neutron capture on ^{56}Fe present in the source holder was used also. The deviations from linearity of the γ peak positions were less than 30 keV over the whole range. The space correction allows one to equalize the charge throughout the detector with an accuracy of better than 3%. The energy resolution scales approximately as $(\sigma/E) \simeq (0.058 + 1.1 \times 10^{-3}E)/\sqrt{E}$, where E is given in MeV (Fig. 2).

The position of an event is determined by using a photon time-of-flight reconstruction algorithm. The resolution in the event position reconstruction is 13 ± 2 cm in the x and y coordinates and 14 ± 2 cm in z (vertical axis), measured with the ^{214}Bi-^{214}Po β–α decay sequence. The spatial resolution is expected to scale as $N^{1/2}$, where N is the number of detected photoelectrons.

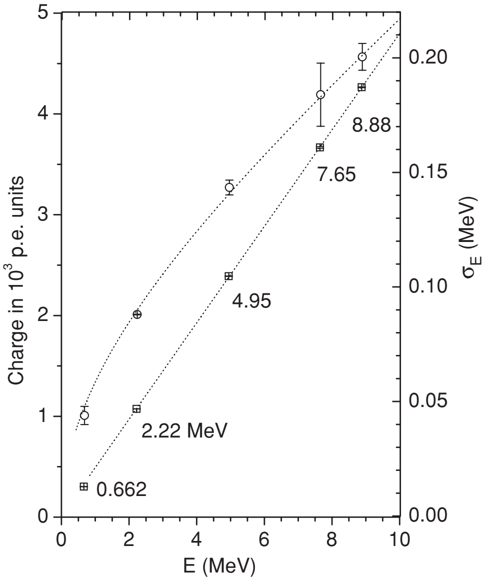

FIG. 2. The dependency of registered charge vs energy of γ quanta (squares, left scale). The corresponding energy resolution (σ_E) is indicated on the right scale (circles).

III. DATA ANALYSIS

A. Theoretical considerations

The non-Paulian transitions were searched for in ^{12}C nuclei of the PC. The nucleon level scheme of ^{12}C in a simple shell model is shown in Fig. 3. The non-Paulian transitions that have been searched for in the analysis described in this paper are schematically illustrated. The transition of a nucleon from the P shell to the filled S shell will result in excited non-Paulian nuclei $^{12}\widetilde{\mathrm{C}}$. The excitation energy corresponds to the difference of the binding energies of nucleons on S and P shells and

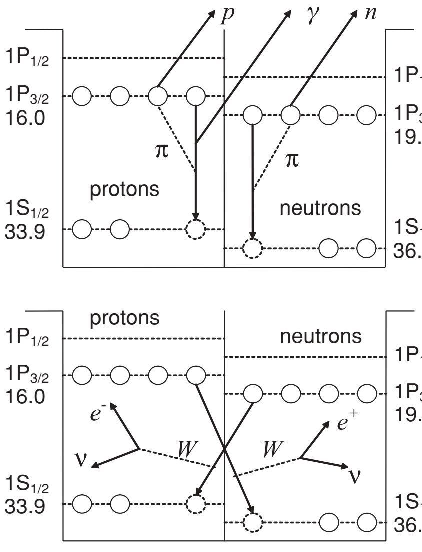

FIG. 3. Occupation of energy levels by protons and neutrons for the ^{12}C ground state in a simple shell model. Schemes of non-Paulian transitions of nucleons from the P shell to the filled S shell (top) with γ, n, p, and α emission and (bottom) with β^{+} and β^{-} emission.

is comparable with the separation energies of protons, S_p, neutrons, S_n, and α particles, S_α. Hence, together with the emission of γ quanta, the emission of n, p, and α is possible. In this paper we also discuss weak processes violating PEP, such as β^+ and β^- decay to a non-Paulian nucleon in the final $1S_{1/2}$ state.

The energy released in the transitions under consideration is the difference between the binding energies of the final and initial nuclei:

$$\begin{aligned} Q(^{12}\mathrm{C} \to \widetilde{X} + Y) &= M(^{12}\mathrm{C}) - M(\widetilde{X}) - M(Y) \\ &= -E_b(^{12}\mathrm{C}) + E_b(\widetilde{X}) + E_b(Y), \end{aligned} \qquad (1)$$

where $\widetilde{X}$ denotes a non-Paulian nucleus, $Y = \gamma, p, n, d, \alpha$ is the particle or nucleus emitted, and E_b is the corresponding binding energies, which are well known for normal nuclei [56]. The signature of non-Paulian transitions with two particles in the final state is a peak in the experimental spectrum with the width defined by the energy resolution of the detector.

In the case of non-Paulian transitions induced by weak interactions, the $\beta^\pm$ spectra have to be observed. The end-point energy of the β spectrum in the reaction $^{12}\mathrm{C} \to {}^{12}\widetilde{\mathrm{N}} + e^- + \overline{\nu}$ is

$$Q = m_n - m_p - m_e - E_b(^{12}\mathrm{C}) + E_b(^{12}\widetilde{\mathrm{N}}). \qquad (2)$$

A similar equation can be written for non-Paulian transition with β^+ emission, but the registered energy will be shifted by $\approx 2m_e$ owing to positron annihilation quanta.

The binding energy of the non-Paulian nuclei with three neutrons or three protons on the $1S_{1/2}$ shell, $E_b(\widetilde{X})$, can be evaluated by considering the binding energy of normal nuclei, $E_b(X)$, and the difference between the binding energies of nucleons on the $1S_{1/2}$ shell, $E_{n,p}(S_{1/2})$, and the binding energy of the last nucleon, $S_{n,p}(X)$:

$$E_b(\widetilde{X}_{n,p}) \simeq E_b(X) + E_{n,p}(1S_{1/2}) - S_{n,p}(X). \qquad (3)$$

The nucleon binding energies for light nuclei (^{12}C, ^{11}B, and others) were measured while studying (p,2p) and (p,np) proton scattering reactions with 1 GeV energy at the PNPI proton synchrocyclotron [57]. Using these data we calculated the Q values (with errors) for different non-Paulian transitions that are shown in Table I. The details of the calculations can be found in our previous work [50].

For all other reactions such as $^{12}\mathrm{C} \to {}^{10}\widetilde{\mathrm{B}} + d$, $^{12}\mathrm{C} \to {}^{9}\widetilde{\mathrm{B}} + t$, $^{12}\mathrm{C} \to {}^{9}\widetilde{\mathrm{Be}} + {}^{3}\mathrm{He}$, $^{12}\mathrm{C} \to {}^{6}\widetilde{\mathrm{Li}} + {}^{6}\mathrm{Li}$, and $^{12}\mathrm{C} \to {}^{6}\widetilde{\mathrm{Li}} + {}^{4}\mathrm{He} + d$, except in the process $^{12}\mathrm{C} \to {}^{9}\widetilde{\mathrm{B}}_{3p} + t$, the Q values are negative.

TABLE I. The energies released in the transitions with non-Paulian nuclei with three neutrons or three protons on the S shell in the final state.

Channel	Q_{3p} (MeV)	Q_{3n} (MeV)
$^{12}\mathrm{C} \to {}^{12}\widetilde{\mathrm{C}} + \gamma$	17.9 ± 0.9	17.7 ± 0.6
$^{12}\mathrm{C} \to {}^{11}\widetilde{\mathrm{B}} + p$	6.3 ± 0.9	7.8 ± 1.0
$^{12}\mathrm{C} \to {}^{11}\widetilde{\mathrm{C}} + n$	6.5 ± 0.9	4.5 ± 0.6
$^{12}\mathrm{C} \to {}^{8}\widetilde{\mathrm{Be}} + \alpha$	3.0 ± 0.6	2.9 ± 0.9
$^{12}\mathrm{C} \to {}^{12}\widetilde{\mathrm{N}} + e^- + \overline{\nu_e}$	18.9 ± 0.9	–
$^{12}\mathrm{C} \to {}^{12}\widetilde{\mathrm{B}} + e^+ + \nu_e$	–	17.8 ± 0.9

Using the obtained Q values one can calculate the detector response for all the reactions just mentioned. The recoil energy of nuclei and quenching factors for different particles have to be taken into account.

Because of the uncertainties in the non-Paulian nuclei properties, the prediction of the branching ratio for the emission in each of the aforementioned channels has a poor significance. For the case of the neutron disappearance (e.g., invisible decay $n \to 3\nu$) from the $1S_{1/2}$ shell in ^{12}C nuclei, the branching ratio and spectra of the emitted particles were considered in Ref. [58]. For the excitation energy of ^{11}C of 17 MeV these authors found that the branching ratios for p, n, and α emission are of the same order of magnitude and that it is negligible for γ emission. In the present paper we give the separate limits on the probabilities for each of the non-Paulian reactions. Then, we compare the obtained results with the corresponding rates of normal transitions.

B. Data selection

Candidate events are selected by the following criteria: (1) Events must have a unique cluster of PMT hits; (2) events should not be flagged as muons by the outer Cherenkov detector; (3) events should not follow a muon within a time window of 2 ms; (4) events should not be followed by another event within a time window of 2 ms except in case of neutron emission; (5) events must be reconstructed within the detector volume. Depending on the specific channel under study, pulse-shape discrimination has also been applied to select events induced by γ, β, p, or α.

The experimental energy spectra of Borexino in the range (1.0–14) MeV, collected during 485 days of data-taking (live time), is shown in Fig. 4. The raw spectrum is presented at the top. At energies below 3 MeV, the spectrum is dominated

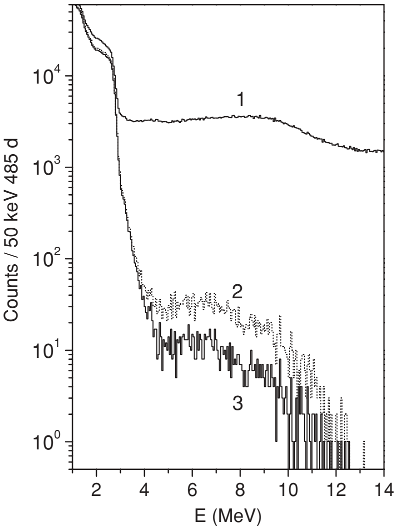

FIG. 4. Energy spectra of the events and effect of the selection cuts. From top to bottom: (1) raw spectrum; (2) with 2-ms muon veto cut; (3) with events within 0.7 s of a muon crossing the SSS removed.

by 2.6-MeV γ rays from the β decay of ^{208}Tl resulting from radioactive contaminations in the PMTs and in the SSS.

The second spectrum is obtained by vetoing all events within 2 ms after the muon. The events were selected with the additional requirement that the mean time of the hits belonging to the cluster with respect to the first hit of the cluster must be $\leqslant$100 ns and the time corresponding to the maximum density of hits must be $\leqslant$30 ns. This cut rejects residual muons that were not tagged by the outer water Cherenkov detector and that interacted in the PC buffer regions. To reduce the background from the short-lived isotopes (^{9}Li, 178 ms; ^{8}He, 119 ms) induced by muons, an additional 0.7-s veto is applied after each muon crossing the SSS (line 3, Fig. 4). This cut induces 3.5% dead time that reduces the live time to 467.8 days. No events with energy higher than 12.5 MeV passed this cut. This fact will be used to set limits on the PEP-forbidden transitions with γ and $\beta^{\pm}$ emissions that have large Q values (see Table I).

For PEP-forbidden transitions with nucleon emission we analyzed the data in the range 0.5–8.0 MeV. In this energy region it is necessary to apply a fiducial volume (FV) cut in addition to the cuts already described to reject external background. Figure 5 shows the effect of selecting only the innermost 100 tons of scintillator by applying a cut $R = 3.02$ m (line 1).

The spectrum below 3 MeV is significantly suppressed by the fiducial cut, by a factor of $\approx 10^2$. The shape of the background in the range of 1–2 MeV is determined by cosmogenic ^{11}C β^+ decays. In the next stage of data selection we removed couples of correlated events falling in a time window of 2 ms (line 2, Fig. 5). This cut mainly rejects ^{214}Bi-^{214}Po coincidences from the ^{238}U chain.

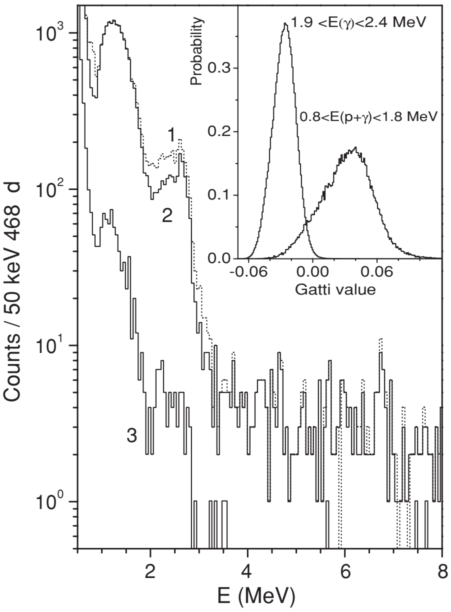

FIG. 5. The energy spectra of events registered inside the FV ($R \leqslant 3.02$ m). Spectrum (1) obtained with 2 ms and 0.7-s muon veto cut; (2) obtained with pairs of correlated events (with time interval $\Delta t \leqslant 2$ ms between signals) removed; (3) of the events with positive Gatti variable. In the inset the values of the Gatti variable obtained with an ^{241}Am-^{9}Be source for protons and 2.22 MeV γ are shown.

Finally, a pulse-shape discrimination analysis based on the Gatti optimal filter [59] is performed to select nucleons. The Gatti parameter is obtained through the following weighted sum: $G = \Sigma P_i S_i$, where S_i is the number of photoelectrons emitted in the time interval $\triangle t_i$. The corresponding weights P_i are computed for average pulse shapes of the signals produced by α and β particles. The parameter G is distributed around a mean value, which is positive for α particles (and protons) and negative for electrons. Line 3 of Fig. 5 shows the events corresponding to positive values of variable G (see Refs. [54,60] for more details).

IV. RESULTS

A. Limits on non-Paulian transitions with emission of γ: $^{12}\mathrm{C} \rightarrow {}^{12}\widetilde{\mathrm{C}} + \gamma$

The limit on the probability of the forbidden transitions $^{12}\mathrm{C} \rightarrow {}^{12}\widetilde{\mathrm{C}} + \gamma$ violating the PEP is based on the experimental fact that no events above 12.5 MeV survive the selection cuts.

The lower limits on the lifetime for PEP-violating transitions of nucleons from the P shell to the occupied $1S_{1/2}$ shell were obtained using the formula

$$\tau \geqslant \varepsilon(\Delta E)\frac{N_N N_n}{S_{\mathrm{lim}}}T, \qquad (4)$$

where $\varepsilon(\Delta E)$ is the detection efficiency of an event in the energy interval ΔE, N_N is the number of nuclei under consideration, N_n is the number of nucleons (n and/or p) in the nuclei for which the non-Paulian transitions are possible, T is the total time of measurements, and S_{lim} is the upper limit on the number of candidate events registered in the ΔE energy interval and corresponding to the chosen confidence level.

The total mass of scintillator ($\sim N_N$) and live time (T) are known within 0.2%. The systematic uncertainties of two other parameters [$\varepsilon(\Delta E)$ and S_{lim}] are significantly larger because they depend on the most poorly defined Q values. As a result the lifetime limits (τ) are calculated for the conservative Q values.

As shown in Table I, the most probable energy of γ quanta emitted in the nucleon transition from the shell $1P_{3/2}$ to the shell $1S_{1/2}$ is $\simeq$17.8 MeV. By taking into account the error of Q values, the energy of γ quanta is inside the energy interval 16.4–19.4 MeV with 90% probability. The efficiency of γ detection is found for the conservative value $E_\gamma = 16.4$ MeV. The response function of the Borexino detector to the γ rays of this energy was found by Monte Carlo (MC) simulations based on the GEANT4 code. The uniformly distributed γ rays were simulated inside the inner vessel (PC + PPO) and in the 1-m-thick layer of buffer (PC + DMP) surrounding the inner vessel. The response function is shown in Fig. 6; the obtained efficiency of 16.4-MeV γ detection is $\varepsilon_{\Delta E} = 0.50$.

The number of ^{12}C target nuclei in 533 tons of PC is $N_N = 2.37 \times 10^{31}$ (which is found by taking into account the isotopic abundance of ^{12}C). The number of nucleons on the P shell is $N_n = 8$, the total data-taking time is $T = 1.282$ yr, and the upper limit on the number of candidate events is $S_{\mathrm{lim}} = 2.44$

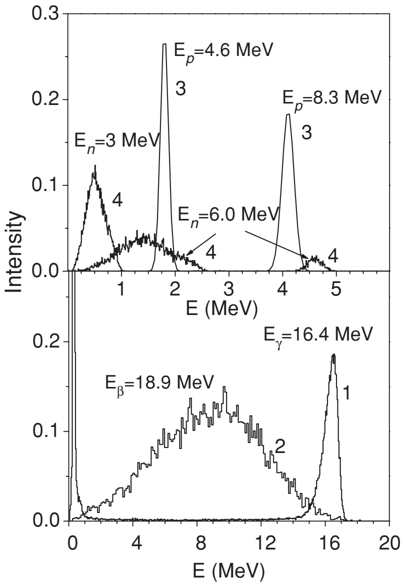

FIG. 6. The response functions of Borexino: (1) $^{12}\mathrm{C} \to {}^{12}\widetilde{\mathrm{C}} + \gamma$ (16.4 MeV) decays in IV and 1-m thick-layer of buffer; (2) $^{12}\mathrm{C} \to {}^{12}\widetilde{\mathrm{N}} + e^- + \overline{\nu}$ (18.9 MeV); (3) $^{12}\mathrm{C} \to {}^{11}\widetilde{\mathrm{B}} + p$ (4.6 and 8.3 MeV); (4)$^{12}\mathrm{C} \to {}^{11}\widetilde{\mathrm{C}} + n$ (3.0 and 6.0 MeV).

with 90% C.L. in accordance with the Feldman-Cousins procedure [61]. The limit obtained using the cited numbers is

$$\tau_\gamma(^{12}\mathrm{C} \to {}^{12}\widetilde{\mathrm{C}} + \gamma) \geqslant 5.0 \times 10^{31}\ \mathrm{yr}, \qquad (5)$$

for the 90% C.L. The result improves by more than four orders of magnitude our previous limit, obtained with CTF [50]: $\tau(^{12}\mathrm{C} \to {}^{12}\widetilde{\mathrm{C}} + \gamma) \geqslant 2.1 \times 10^{27}$ yr. This result is stronger than the one obtained with the NEMO-2 detector, $\tau(^{12}\mathrm{C} \to {}^{12}\widetilde{\mathrm{C}} + \gamma) \geqslant 4.2 \times 10^{24}$ yr [44], and is comparable with that from the Kamiokande detector for ^{16}O nuclei, $\tau(^{16}\mathrm{O} \to {}^{16}\widetilde{\mathrm{O}} + \gamma) \geqslant 1.0 \times 10^{32}$ yr for γ rays with energies of 19–50 MeV [43].

The limit on the total lifetime of nucleons can be found from the limits on τ_γ as $\tau = \tau_\gamma \mathrm{Br}(\gamma)$, where $\mathrm{Br}(\gamma) = \Gamma_\gamma / \Gamma_{\mathrm{tot}}$ is the branching fraction of γ decay. For the case of the ^{16}O nucleus the calculated value of $\mathrm{Br}(\gamma)$ is in the interval $(2.7–10.4) \times 10^{-5}$ [43]. Unlike the Kamiokande, the Borexino can directly detect the non-Paulian transitions with p, n, or α emission.

B. Limits on non-Paulian transitions in ^{12}C with proton emission $^{12}\mathrm{C} \to {}^{11}\widetilde{\mathrm{B}} + p$

Using the data of Table I, one can obtain that the energy released in these transition is within the 5.0–9.0 MeV interval with a probability of 90%. By taking into account the recoil energy of the $^{11}\widetilde{\mathrm{B}}$ nucleus, the energy of the proton is 4.6–8.3 MeV.

The response function of protons was simulated by an MC code that takes into account the quenching factor for protons (Fig. 6). The empirical Birks' constant [55] was determined from the spectrum of recoil protons measured with an ^{241}Am-^{9}Be source. It was found that the light yield for a proton with an energy $E_p = 4.6(8.3)$ MeV corresponds to an electron energy of $E_e = 1.8(4.1)$ MeV. This means that the proton peak can be found in the energy interval 1.8–4.1 MeV with 90% probability. The uncertainty of the peak position is much higher than the energy resolution of the detector ($\sigma_E \cong$ 80 keV for $E_e = 2$ MeV). First, we looked for the proton's peak in the spectrum of single events obtained with the FV cut (line 2, Fig. 5). The measured spectrum is fitted by a polynomial function and a Gaussian for the proton peak with different positions. Except for the region of the 2.614-MeV γ peak, this procedure gives $S_{\mathrm{lim}} = 52$ at 90% C.L. The lower limit on the lifetime was found from the formula (4) by taking into account that $N_N = 4.45 \times 10^{30}$ for 100 tons FV mass:

$$\tau_p(^{12}\mathrm{C} \to {}^{11}\widetilde{\mathrm{B}} + p) \geqslant 8.9 \times 10^{29}\ \mathrm{yr}\ (90\%\ \mathrm{C.L.}). \qquad (6)$$

The more stringent limit can be obtained by analyzing the spectrum of signals with positive values of the Gatti variable (line 3, Fig. 5), which correspond to the detection of α particles or protons. The lower limit on the lifetime is

$$\tau_p(^{12}\mathrm{C} \to {}^{11}\widetilde{\mathrm{B}} + p) \geqslant 2.1 \times 10^{30}\ \mathrm{yr}\ (90\%\ \mathrm{C.L.}), \qquad (7)$$

where the efficiency of the Gatti cut, $\varepsilon = 0.89$, was taken into account. It is worth noting that the systematic error for 100 tons FV mass defined by a software cut is 6%. This value is estimated on the basis of the distribution of reconstructed vertices of uniform background sources [53].

The upper limits on nuclear instabilities of the ^{12}C nucleus differ from the limits (6) and (7) by a factor $N_n = 8$. They are about four orders of magnitude stronger than the ones obtained with the 300-kg NaI ELEGANT V detector, $\tau(^{23}\mathrm{Na}, {}^{127}\mathrm{I} \to {}^{22}\widetilde{\mathrm{Ne}}, {}^{126}\widetilde{\mathrm{Te}} + p) \geqslant 1.7 \times 10^{25}$ yr (90% C.L.) for protons with $E_p \geqslant$18 MeV [45], and with the 250-kg NaI DAMA/LIBRA detector, $\tau(^{23}\mathrm{Na}, {}^{127}\mathrm{I} \to {}^{22}\widetilde{\mathrm{Ne}}, {}^{127}\widetilde{\mathrm{Te}} + p) \geqslant 1.9 \times 10^{25}$ yr (90% C.L.) for protons with $E_p \geqslant 10$ MeV [35].

The energy of α particles emitted in $^{12}\mathrm{C} \to {}^{8}\widetilde{\mathrm{Be}} + \alpha$ decay can be found in the 1.0–3.0 MeV interval. Because of the quenching factor, this corresponds to an electron energy range of 70–250 keV. Because an energy of 70 keV is close to the Borexino lower energy threshold we have not analyzed this reaction with the Borexino data. Our limit on this mode of transition, which was obtained using the CTF measurements with 20-keV threshold, is $\tau(^{12}\mathrm{C} \to {}^{8}\widetilde{\mathrm{Be}} + \alpha) \geqslant 6.1 \times 10^{23}$ yr (90% C.L.).

C. Limit on non-Paulian transition in ^{12}C with neutron emission: $^{12}\mathrm{C} \to {}^{11}\widetilde{\mathrm{C}} + n$

Following the calculations of the previous section, one can obtain that the kinetic energy of the initial neutron is in the 3.2–7.3 MeV interval with 90% probability. The resulting neutrons are thermalized in the hydrogen-rich media of the organic scintillator. The lifetime of neutrons in PC is $\tau \cong$ 250 μs, after which they are captured by protons. The cross section for the capture on a proton for a thermal neutron is 0.33 b. The capture of thermal neutrons via $n + p \to d + \gamma$ is followed by γ emission with an energy of 2.2 MeV. The cross sections are much smaller for capture on ^{12}C nuclei ($\sigma_\gamma = 3.5$ mb, $E_\gamma = 4.95$ MeV). As a result, the 4.95-MeV peak intensity is about 1% of that of the 2.2-MeV peak (Fig. 1).

The background levels measured in Borexino at 2.2 MeV energy can be used to obtain an upper limit on the number of γ rays with 2.2 MeV energy, and as a result, a limit on the probability of neutron production in the reactions $^{12}\mathrm{C}\to\,^{11}\widetilde{\mathrm{C}}+n$. Because protons that were scattered during the thermalization can be registered by the detector the sequential events were not cut out in the data selection (see Fig. 5, line 1). The response function of the Borexino to the 2.2-MeV γ rays was precisely measured with an $^{241}\mathrm{Am}$-$^{9}\mathrm{Be}$ neutron source. The position and width of the peak are well known; the fitting procedure gives $S_{\mathrm{lim}}=57$. Using Eq. (4) one can obtain the limit on the probability for neutron emission: $\tau_n(^{12}\mathrm{C}\to\,^{11}\widetilde{\mathrm{C}}+n)\geqslant 8.1\times10^{29}$ yr (90% C.L.)

More stringent limits can be obtained by selecting two consequential events inside the full PC volume, the first signal being from the recoil protons and the second one from the 2.2-MeV γ ray from the neutron capture. Candidate events were searched among all the correlated events occurring within 1.25 ms (5τ) one after another, excluding coincidence times smaller than 20 μs. The energy of the prompt event was set to be $E\geqslant0.5$ MeV. The lower threshold is defined by the minimal neutron energy of 3.2 MeV (visible energy of 0.6 MeV) by taking into account the rate of random coincidences. The response functions for neutrons with energies of 3.0 and 6.0 MeV are shown in Fig. 6. The energy of the second event was required to be $1.0\leqslant E\leqslant2.4$ MeV for detecting the 2.2-MeV γ rays with high efficiency. Additionally, the restored positions of the events have to be within 2 m distance owing to the high energy of the initial neutron. In such a way, 52 events were selected. Then for different neutron energies E_n inside a 3.2–7.3 MeV interval, the corresponding energy regions for recoil proton signals were calculated (see lines 4, Fig. 6). If E_n exceeds the energy of the first exited state of $^{12}\mathrm{C}$ then the high-energy part connected with detection of 4.44-MeV γ rays appears in the spectrum of the prompt events. The maximal value of the correlated events, $N=26$, was found for the ranges of 0.6–2.3 and 4.3–5.0 MeV that correspond to 6-MeV neutrons. By taking into account the probability of finding a 6.0-MeV neutron signal in these ranges ($\varepsilon=0.9$), the efficiency of registering 2.2-MeV γ rays ($\varepsilon=0.96$), the full number of $^{12}\mathrm{C}$ atoms in the inner vessel, $N_N=1.24\times10^{31}$, and $S_{\mathrm{lim}}=33$ for 90% C.L., the limit is

$$\tau_n(^{12}\mathrm{C}\to\,^{11}\widetilde{\mathrm{C}}+n)\geqslant 3.4\times10^{30}\ \mathrm{yr}\ (90\%\ \mathrm{C.L.}). \tag{8}$$

This result is eight orders of magnitude stronger than the one obtained through searching for spontaneous neutron emission from lead: $\tau(\mathrm{Pb}\to\widetilde{\mathrm{Pb}}+n)\geqslant2.1\times10^{22}$ yr (68% C.L.) [47].

D. Limits on non-Paulian $\beta^{\pm}$ transitions: $^{12}\mathrm{C}\to\,^{12}\widetilde{\mathrm{N}}+e^-+\overline{\nu}$ and $^{12}\mathrm{C}\to\,^{12}\widetilde{\mathrm{B}}+e^++\nu$

The energy released in the reaction $^{12}\mathrm{C}\to\,^{12}\widetilde{\mathrm{N}}+e^-+\overline{\nu}$ is in an interval of 16.4–21.4 MeV. The shape of the β^- spectrum with the most probable end-point energy of 18.9 MeV is shown in Fig. 6. The spectrum was determined by an MC method. The limit on the probability of non-Paulian β^- transition was based again on the fact of observing no events with $E_e\geqslant12.5$ MeV not accompanied by a muon veto signal. The obtained efficiency of detection of electrons with energies $E_e>12.5$ MeV is $\varepsilon=0.12$. The limit on the lifetime of neutrons ($N_n=4$) in $^{12}\mathrm{C}$ with respect to the transitions violating the PEP is

$$\tau_{\beta^-}(^{12}\mathrm{C}\to\,^{12}\widetilde{\mathrm{N}}+e^-+\overline{\nu})\geqslant3.1\times10^{30}\ \mathrm{yr}\ (90\%\ \mathrm{C.L.}). \tag{9}$$

This result is six orders of magnitude stronger than the one obtained by NEMO-2, $\tau(^{12}\mathrm{C}\to\,^{12}\widetilde{\mathrm{N}}+e^-+\overline{\nu})\geqslant3.1\times10^{24}$ yr (90% C.L.) [44].

The data available from the LSD detector [62] situated in the tunnel under Mont Blanc allow us to obtain a qualitative limit for this decay mode. In Ref. [48], it is claimed that only two events were observed with energies higher than 12 MeV during 75 days of data-taking with the detector loaded with 7.2 tons of scintillator, containing 3×10^{29} $^{12}\mathrm{C}$ nuclei. The upper limit that can be obtained using formula (4) with these data [with $S_{\mathrm{lim}}=5.91$ events for 90% C.L. and detection efficiency $\varepsilon(E\geqslant12\ \mathrm{MeV})=0.23$] is $\tau(^{12}\mathrm{C}\to\,^{12}\widetilde{\mathrm{N}}+e^-+\overline{\nu})\geqslant9.5\times10^{27}$ yr (90% C.L.).

The end-point energy of the β^+ spectrum is 16.8 MeV, but the spectrum is shifted toward higher energies by $\simeq$0.85 MeV by the registering of annihilation quanta (Fig. 6). The efficiency of the $^{12}\mathrm{C}\to\,^{12}\widetilde{\mathrm{B}}+e^++\nu$ transition detection with energy release $E>12.5$ MeV is $\varepsilon=0.079$. The lower limit on the lifetime of the proton in the $^{12}\mathrm{C}$ nuclei is then

$$\tau_{\beta^+}(^{12}\mathrm{C}\to\,^{12}\widetilde{\mathrm{B}}+e^++\nu)\geqslant2.1\times10^{30}\ \mathrm{yr}\ (90\%\ \mathrm{C.L.}). \tag{10}$$

The limits obtained by the NEMO-2 Collaboration for this reaction are six orders of magnitude weaker: $\tau(^{12}\mathrm{C}\to\,^{12}\widetilde{\mathrm{B}}+e^++\nu)\geqslant2.6\times10^{24}$ yr (90% C.L.) [44].

The final limits on the nucleon instability are shown in Table II in comparison with the the previous results obtained for the same PEP-violating transitions. The limit [35] relates to the instability of $^{23}\mathrm{Na}$ and $^{127}\mathrm{I}$ nuclei; all other limits are given per nucleon for which the non-Paulian transition is possible.

E. Limits on the relative strength of non-Paulian transitions

The PEP-forbidden transitions with emission of γ, n or p, and (e,ν) pairs can be induced by electromagnetic, strong, and weak interactions, correspondingly. The obtained upper limits on lifetime for different processes can be converted to limits on the relative strength of non-Paulian transitions to the normal one: $\delta^2=\widetilde{\lambda}/\lambda$, where $\lambda=1/\tau$ is unit time probability (rate) of forbidden ($\widetilde{\lambda}$) and normal (λ) transitions. The ratio $\delta^2=(g_{\mathrm{PV}}/g_{\mathrm{NT}})^2$ is a measure of the violation of

TABLE II. Mean lifetime limits for non-Paulian transitions of nucleons in the Borexino.

Channel	τ_{lim} (yr) 90% C.L.	Previous limits	Ref.
$^{12}\mathrm{C}\to\,^{12}\widetilde{\mathrm{C}}+\gamma$	5.0×10^{31}	$4.2\times10^{24}(^{12}\mathrm{C})$	[44]
		$1.0\times10^{32}(^{16}\mathrm{O})$	[43]
$^{12}\mathrm{C}\to\,^{11}\widetilde{\mathrm{B}}+p$	8.9×10^{29}	$1.9\times10^{25}(^{23}\mathrm{Na},^{127}\mathrm{I})$	[35]
$^{12}\mathrm{C}\to\,^{11}\widetilde{\mathrm{C}}+n$	3.4×10^{30}	$2.1\times10^{22}(^{\mathrm{nat}}\mathrm{Pb})$	[47]
$^{12}\mathrm{C}\to\,^{12}\widetilde{\mathrm{N}}+e^-+\overline{\nu_e}$	3.1×10^{30}	$9.5\times10^{27}(^{12}\mathrm{C})$	[48,62]
$^{12}\mathrm{C}\to\,^{12}\widetilde{\mathrm{B}}+e^++\nu_e$	2.1×10^{30}	$2.6\times10^{24}(^{12}\mathrm{C})$	[44]

the PEP and represents the mixing probability of nonfermion statistics allowing the transitions to the occupied states. In particular, in the quon model of PEP violation [9,10] the parameter $\delta^2 = \beta^2/2$ corresponds to the probability of an admixed symmetric component of the particle. In this way one can compare the experimental limits on the lifetime obtained for different nuclei and atoms.

The decay width of the nuclear electric dipole 16.4-MeV $E1$ γ transition from the P to the S shell given by the Weisskopf estimate is $\Gamma_\gamma \approx 1.5$ keV, and the rate of normal $E1$ transition is $\lambda = \Gamma_\gamma/\hbar = 2.3 \times 10^{18}$ s^{-1}. With the obtained upper limit on τ_γ (5), the ratio $\delta^2_\gamma = \widetilde{\lambda}(^{12}\mathrm{C})/\lambda(^{12}\mathrm{C})$ is less than 2.2×10^{-57} (90% C.L.). This limit is close to the Kamiokande detector result for ^{16}O nuclei of $\delta^2_\gamma = 2.3 \times 10^{-57}$ [43].

Although the $E1$ transition is the fastest among the γ transitions, the width of hadron emissions is three to four orders larger than that of γ transitions. The widths of single S-hole states in ^{12}C measured for $(p,2p)$ and (p,pn) reactions are $\Gamma_{n,p} \cong 12$ MeV [57]. As a result, the detection of protons or neutrons gives a more stringent limit on the relative strength of PEP-forbidden transitions than the detection of γ rays if one can set a similar limit on the lifetime for both decays. Using the lower limits on τ_p (7) and τ_n (8) one can obtain the limits $\delta^2_p = \widetilde{\lambda}/\lambda \leqslant 1.6 \times 10^{-59}$ and $\delta^2_n \leqslant 4.1 \times 10^{-60}$ at 90% C.L. This result is more then four orders of magnitude stronger than the one obtained by the DAMA Collaboration [35].

The non-Paulian $\beta^\pm$ transitions are first-order forbidden $P \to S$ transitions (Fig. 3, bottom). The $\log(ft_{1/2})$ values for such transitions equal 7.5 ± 1.5. The conservative value $\log(ft_{1/2}) = 9$ corresponds to the large enough lifetime $\tau \approx 480$ s for $Q = 18.9$ MeV in the case of β^- decay (where the level width is $\Gamma_{\beta^-} \approx 1.4 \times 10^{-18}$ eV) and $\tau \approx 1050$ s ($Q = 17.8$ MeV, β^+). As a result, the restrictions on the relative strength of non-Paulian $\beta^\pm$ decays are significantly weaker than restrictions on δ^2_γ and $\delta^2_{p,n}$: $\delta^2_{\beta^-} \leqslant 2.1 \times 10^{-35}$ and $\delta^2_{\beta^+} \leqslant 6.4 \times 10^{-35}$ (90% C.L.). The strongest previous result ($\delta^2_{\beta^-} \leqslant 6.5 \times 10^{-34}$) was obtained in Ref. [48] with LSD data [62].

Although the limits on the relative strength of $\beta^\pm$ transitions are more than 20 orders of magnitude weaker than the limits on the relative strengths of non-Paulian transitions with p, n, and γ emission (Fig. 3, top), there is a significant difference between these processes. It was mentioned befor that a new particle (p or n) arises in a non-Paulian state when $\beta^\pm$ decay occurs; thus Amado-Primakoff arguments for identical particles may not be valid [13,48]. In this way the limit on $\delta^2_{\beta^\pm}$ can be compared with the similar limit obtained by the VIP experiment: $\delta^2 = \beta^2/2 \leqslant 4.5 \times 10^{-28}$ [38].

TABLE III. Upper limits on the relative strength, $\delta^2 = \widetilde{\lambda}/\lambda$ (at 90% C.L.), for non-Paulian transitions in the Borexino.

Decay	$\widetilde{\lambda}(^{12}\mathrm{C})$ (s^{-1})	$\lambda(^{12}\mathrm{C})$ (s^{-1})	$\delta^2 = \widetilde{\lambda}/\lambda$	Previous limits	Ref.
γ	5.0×10^{-39}	2.3×10^{18}	2.2×10^{-57}	2.3×10^{-57}	[43]
N(n,p)	7.4×10^{-38}	1.8×10^{22}	4.1×10^{-60}	3.5×10^{-55}	[35]
(e,ν)	4.1×10^{-38}	2.0×10^{-3}	2.1×10^{-35}	6.5×10^{-34}	[48,62]

The upper limits obtained on the relative strengths of non-Paulian transitions are shown in Table III. For transitions with (n,p) and $\beta^\pm$ emission the stronger limit is included.

V. CONCLUSIONS

Using the unique features of the Borexino detector—extremely low background, large scintillator mass of 278 tons, low energy threshold, and a carefully designed muon-veto system—the following new limits on non-Paulian transitions of nucleons from the $1P_{3/2}$ shell to the $1S_{1/2}$ shell in ^{12}C with the emission of γ, n, p, and $\beta^\pm$ particles have been obtained:

$$\tau(^{12}\mathrm{C} \to {}^{12}\widetilde{\mathrm{C}} + \gamma) \geqslant 5.0 \times 10^{31}\ \mathrm{yr},$$

$$\tau(^{12}\mathrm{C} \to {}^{11}\widetilde{\mathrm{B}} + p) \geqslant 8.9 \times 10^{29}\ \mathrm{yr},$$

$$\tau(^{12}\mathrm{C} \to {}^{11}\widetilde{\mathrm{C}} + n) \geqslant 3.4 \times 10^{30}\ \mathrm{yr},$$

$$\tau(^{12}\mathrm{C} \to {}^{12}\widetilde{\mathrm{N}} + e^- + \nu) \geqslant 3.1 \times 10^{30}\ \mathrm{yr},$$

and

$$\tau(^{12}\mathrm{C} \to {}^{12}\widetilde{\mathrm{B}} + \mathrm{e}^+ + \overline{\nu}) \geqslant 2.1 \times 10^{30}\ \mathrm{yr},$$

all with 90% C.L.

Comparing these values with the data of Table II, one can see that these limits for non-Paulian transitions in ^{12}C with γ, p, n, and $\beta^\pm$ emissions are the best to date. The obtained lifetime limits allow us to introduce the new upper limits on the relative strengths of the non-Paulian transitions to the normal ones: $\delta^2_\gamma \leqslant 2.2 \times 10^{-57}$, $\delta^2_N \leqslant 4.1 \times 10^{-60}$, and $\delta^2_\beta \leqslant 2.1 \times 10^{-35}$, all at 90% C.L.

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