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

G. Bellini¹, J. Benziger², S. Bonetti¹, M. Buizza Avanzini¹, B. Caccianiga¹, L. Cadonati³, F. Calaprice⁴, C. Carraro⁵, A. Chavarria⁴, F. Dalnoki-Veress⁴, D. D'Angelo¹, S. Davini⁵, H. de Kerret⁶, A. Derbin⁷, A. Etenko⁸, K. Fomenko⁹, D. Franco¹, C. Galbiati⁴, S. Gazzana¹⁰, C. Ghiano¹⁰, M. Giammarchi¹, M. Goeger-Neff¹¹, A. Goretti⁴, C. Grieb¹², E. Guardincerri⁵, S. Hardy¹², Aldo Ianni¹⁰, Andrea Ianni⁴, M. Joyce¹², G. Korga¹⁰, D. Kryn⁶, M. Leung⁴, T. Lewke¹¹, E. Litvinovich⁸, B. Loer⁴, P. Lombardi¹, L. Ludhova¹, I. Machulin⁸, S. Manecki¹², W. Maneschg¹³, G. Manuzio⁵, Q. Meindl¹¹, E. Meroni¹, L. Miramonti¹, M. Misiasek^{15,10}, D. Montanari^{10,4}, V. Muratova⁷, L. Oberauer¹¹, M. Obolensky⁶, F. Ortica¹⁴, M. Pallavicini⁵, L. Papp¹², L. Perasso¹, S. Perasso⁵, A. Pocar³, R.S. Raghavan¹², G. Ranucci¹, A. Razeto¹⁰, A. Re¹, P. Risso⁵, A. Romani¹⁴, D. Rountree¹², A. Sabelnikov⁸, R. Saldanha⁴, C. Salvo⁵, S. Schoenert¹³, H. Simgen¹³, M. Skorokhvatov⁸, O. Smirnov⁹, A. Sotnikov⁹, S. Sukhotin⁸, Y. Suvorov^{1,8}, R. Tartaglia¹⁰, G. Testera⁵, D. Vignaud⁶, R.B. Vogelaar¹², F. von Feilitzsch¹¹, Y. Winter¹¹, M. Wojcik¹⁵, M. Wurm¹¹, J. Xu⁴, O. Zaimidoroga⁹, S. Zavatarelli⁵, G. Zuzel¹³

¹ Dipartimento di Fisica, Universita' degli Studi e INFN, 20133 Milano, Italy

² Chemical Engineering Department, Princeton University, Princeton, NJ 08544, USA

³ Physics Department, University of Massachusetts, Amherst, AM01003, USA

⁴ Physics Department, Princeton University, Princeton, NJ 08544, USA

⁵ Dipartimento di Fisica, Universita' e INFN, Genova 16146, Italy

⁶ Laboratoire AstroParticule et Cosmologie, 75231 Paris cedex 13, France

⁷ St. Petersburg Nuclear Physics Institute, 188350 Gatchina, Russia

⁸ RRC Kurchatov Institute, 123182 Moscow, Russia

⁹ Joint Institute for Nuclear Research, 141980 Dubna, Russia

¹⁰ INFN Laboratori Nazionali del Gran Sasso, SS 17 bis Km 18+910, 67010 Assergi (AQ), Italy

¹¹ Physik Department, Technische Universitaet Muenchen, 85747 Garching, Germany

¹² Physics Department, Virginia Polytechnic Institute and State University, Blacksburg, VA 24061, USA

¹³ Max-Planck-Institut fuer Kernphysik, 69029 Heidelberg, Germany

¹⁴ Dipartimento di Chimica, Universita' e INFN, 06123 Perugia, Italy

¹⁵ M. Smoluchowski Institute of Physics, Jagellonian University, 30059 Krakow, Poland

Borexino collaboration

Abstract. 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 β^\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 t) of the Borexino detector, the following most stringent up-to-date experimental bounds on PEP violating transitions of nucleons have been established: $\tau(^{12}\text{C} \rightarrow ^{12}\tilde{\text{C}} + \gamma) \geq 5.0 \cdot 10^{31}$ y, $\tau(^{12}\text{C} \rightarrow ^{11}\tilde{\text{B}} + p) \geq 8.9 \cdot 10^{29}$ y, $\tau(^{12}\text{C} \rightarrow ^{11}\tilde{\text{C}} + n) \geq 3.4 \cdot 10^{30}$ y, $\tau(^{12}\text{C} \rightarrow ^{12}\tilde{\text{N}} + e^- + \tilde{\nu}_e) \geq 3.1 \cdot 10^{30}$ y and $\tau(^{12}\text{C} \rightarrow ^{12}\tilde{\text{B}} + e^+ + \nu_e) \geq 2.1 \cdot 10^{30}$ y, 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: $\delta_\gamma^2 \leq 2.2 \cdot 10^{-57}$, $\delta_N^2 \leq 4.1 \cdot 10^{-60}$ and $\delta_\beta^2 \leq 2.1 \cdot 10^{-35}$.

Key words. Pauli exclusion principle – low background measurements

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1 Introduction

The exclusion principle was formulated by W. Pauli in 1925 and in its original form postulated that " these 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 anti-symmetric 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 anti-commutativity of the fermion creation and annihilation operators.

Although 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].

The experimental searches for the 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].

In 1987-91 theoretical models implicating PEP violation were constructed by Ignatiev and Kuzmin [7], Greenberg and Mohapatra [8, 9, 10] and by 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 non-contradictory "small" parameter, as in the case of P - and CP -symmetry violation or L - and B - non-conservation. 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 [2, 14, 15]. More reviews and references can be found in [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 et al. [18, 19] and Nolte et al. [20] who looked for non-Paulian exotic atoms of ^{20}Ne and ^{36}Ar with 3 electrons on K-shell using mass spectroscopy on fluorine and chlorine samples. Similarly, the atoms of Be with 4 electrons in 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 in [22] (Barabash et al.). The PEP-forbidden nuclei of ^5Li with 3 protons in the 1S-shell was searched for by Nolte et al., using time-of-flight mass spectroscopy [23].

Goldhaber was the first to point out that the same experimental data which 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 due to 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 β^\pm -transitions) and can be evaded in composite models of electron or models including extra dimensions [8, 36].

The new method was realized by Ramberg and Snow which 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 3 electrons in the K-shell is $1.7 \cdot 10^{-26}$. 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 ^4He atoms [39] (Deilamian et al.) and molecules of O_2 [40, 41] (Hilborn et al., Angelis et al.) and CO_2 [42] (Modugno et al.).

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 [45, 46, 35] (Elegant-V, DAMA/LIBRA) and n -emission [47] (Koshimoto et al.), non-Paulian β^+ - and β^- - decays [48, 44], (LSD, Kekez et al., NEMO-II) and in nuclear (p, p), (p, α)- reactions on ^{12}C [49] (Miljani et al.).

The strongest limits for non-Paulian transitions in ^{12}C with γ -, p -, n -, α -, and β^\pm - emissions were obtained with a prototype of the Borexino detector - Counting Test Facility (CTF) [50]. In this letter 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 3-4 orders of magnitude with respect to CTF.

2 Experimental set-up and measurements

2.1 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 ultra-pure liquid scintillator. The extremely high radiopurity of the detector and its large mass allow to simultaneously address other fundamental questions particle physics and astrophysics.

The main features of the Borexino detector and its components have been thoroughly described in [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/liter of PPO ($\text{C}_{15}\text{H}_{11}\text{NO}$). 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 (dymethylphthalate-DMP) to reduce their scintillation. The two PC buffers are separated by a second thin nylon membrane to prevent diffusion of radon coming from PMTs, light concentrators and SSS walls towards the scintillator. The scintillator and buffers are contained in a Stainless Steel Sphere (SSS) with diameter 13.7 m. 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 's 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 Cerenkov muon detector (outer detector) to identify the residual muons crossing the detector.

2.2 Detector calibration. 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 using the total collected light from all PMT's. 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].

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 a ^{241}Am - ^9Be neutron source. Fig.1 shows the spectrum obtained with the source placed at the center of the detector. The reactions $^9\text{Be}(\alpha, n)^{12}\text{C}_{\text{gs}}$ and $^9\text{Be}(\alpha, n)^{12}\text{C}^*$ (4.44 MeV) produce two main neutron groups with energies up to 11 MeV 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 's, while the lower (black) one is that of the delayed signals. The energy scale was determined with the 2.22 MeV and 4.95 MeV γ de-excitations following neutron capture on ^1H 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 (due to 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 -peaks positions was less than 30 keV over the whole range. The energy resolution scales approximately

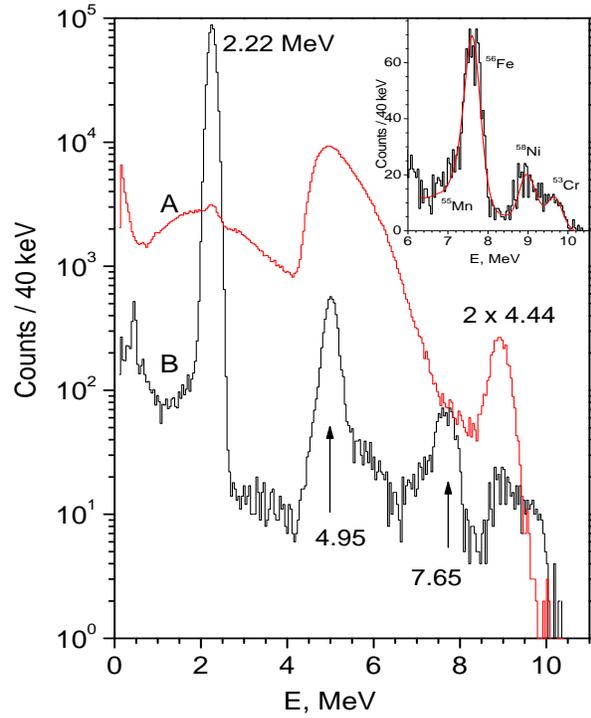


Fig. 1. The energy spectra of prompt (A) and delayed (B) signals registered with ^{241}Am - ^9Be source. In insert, the γ -lines from neutron captures on stainless steel holder of AmBe-source are shown.

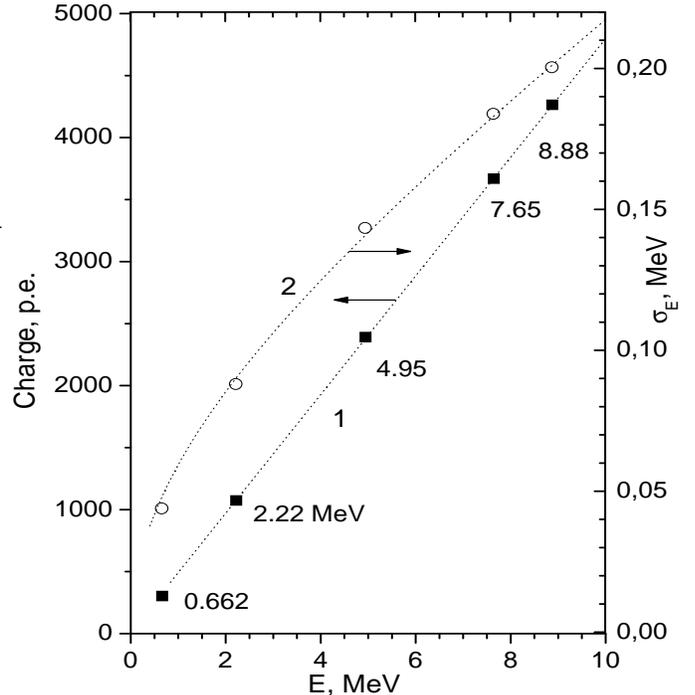


Fig. 2. The dependency of registered charge vs energy of γ -quanta (squares, left scale). The corresponding energy resolution (σ_E) is indicated on the right scale (cycles).

as $(\sigma/E) \simeq (0.058 + 1.1 \cdot 10^{-3}E)/\sqrt{E}$ where E is given in MeV (Fig.2). The position of an event is determined 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, measured with the ^{214}Bi - ^{214}Po $\beta - \alpha$ decay sequence.

3 Data analysis

3.1 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 which 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 $^{12}\tilde{\text{C}}$. The excitation energy corresponds to the difference of the binding energies of nucleons on S- and P-shells and 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, like β^+ - and β^- -decay to a non-Paulian nucleus 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:

$$Q(^{12}\text{C} \rightarrow \tilde{\text{X}} + \text{Y}) = M(^{12}\text{C}) - M(\tilde{\text{X}}) - M(\text{Y}) = -E_b(^{12}\text{C}) + E_b(\tilde{\text{X}}) + E_b(\text{Y}); \quad (1)$$

where $\tilde{\text{X}}$ denotes a non-Paulian nucleus, $\text{Y} = \gamma, p, n, d, \alpha..$ 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 β^\pm -spectra have to be observed. The endpoint energy of the β -spectrum in the reaction $^{12}\text{C} \rightarrow ^{12}\tilde{\text{N}} + e^- + \bar{\nu}$ is

$$Q = m_n - m_p - m_e - E_b(^{12}\text{C}) + E_b(^{12}\tilde{\text{N}}). \quad (2)$$

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

The binding energy of the non-Paulian nuclei with 3 neutrons or 3 protons on the $1S_{1/2}$ -shell $E_b(\tilde{\text{X}})$ can be evaluated 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 $S_{n,p}(X)$:

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

The nucleon binding energies for light nuclei (^{12}C , ^{11}B and others) were measured while studying $(p, 2p)$ and (p, np)

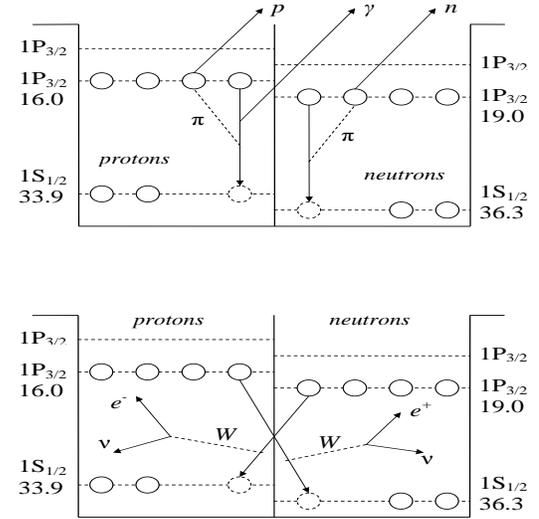


Fig. 3. Occupation of energy levels by protons and neutrons for the ^{12}C ground state in a simple shell model. Schemes of non-Paulian transitions of nucleons from the P-shell to the filled S-shell: a) with γ -, n -, p - and α -emission; b) with β^+ -, β^- -emission.

proton scattering reactions with 1 GeV energy at PNPI proton synchrocyclotron [57]. Using these data we calculated the Q-values (with errors) for different non-Paulian transitions which are shown in Table 1. The details of the calculations can be found in our previous work [50].

Table 1. The energies released in the transitions with non-Paulian nuclei with 3 neutrons or 3 protons on the S-shell in the final state.

Channel	Q_{3p} (MeV)	Q_{3n} (MeV)
$^{12}\text{C} \rightarrow ^{12}\tilde{\text{C}} + \gamma$	17.9 ± 0.9	17.7 ± 0.6
$^{12}\text{C} \rightarrow ^{11}\tilde{\text{B}} + p$	6.3 ± 0.9	7.8 ± 1.0
$^{12}\text{C} \rightarrow ^{11}\tilde{\text{C}} + n$	6.5 ± 0.9	4.5 ± 0.6
$^{12}\text{C} \rightarrow ^8\tilde{\text{Be}} + \alpha$	3.0 ± 0.6	2.9 ± 0.9
$^{12}\text{C} \rightarrow ^{12}\tilde{\text{N}} + e^- + \bar{\nu}_e$	18.9 ± 0.9	-
$^{12}\text{C} \rightarrow ^{12}\tilde{\text{B}} + e^+ + \nu_e$	-	17.8 ± 0.9

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

Using the obtained Q-values one can calculate the detector response for all the reactions mentioned above. 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 above mentioned channels has a

poor significance. For the case of the neutron disappearance (e.g. invisible decay $n \rightarrow 3\nu$) from the $1S_{1/2}$ -shell in ^{12}C nuclei, the branching ratio and spectra of the emitted particles were considered in [59]. For the excitation energy of ^{11}C of 17 MeV they found that the branching ratios for p -, n -, and α -emission are of the same order of magnitude and, 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.

3.2 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 by 2.6 MeV γ 's from the β -decay of ^{208}Tl due to radioactive contaminations in the PMTs and in the SSS.

The second spectrum is obtained by vetoing all events within 2 ms after 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 ≤ 100 ns and the time corresponding to the maximum density of hits must be ≤ 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 due to the short-lived isotopes (^9Li , 178 ms; ^8He , 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 β^\pm - emissions which have large Q-values (see Table 1).

For PEP forbidden transitions with nucleons 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 cut in addition to the cuts described above, in order to reject external background. Fig.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 $\approx 10^2$. The shape of the background in the range of (1÷2) MeV is determined by

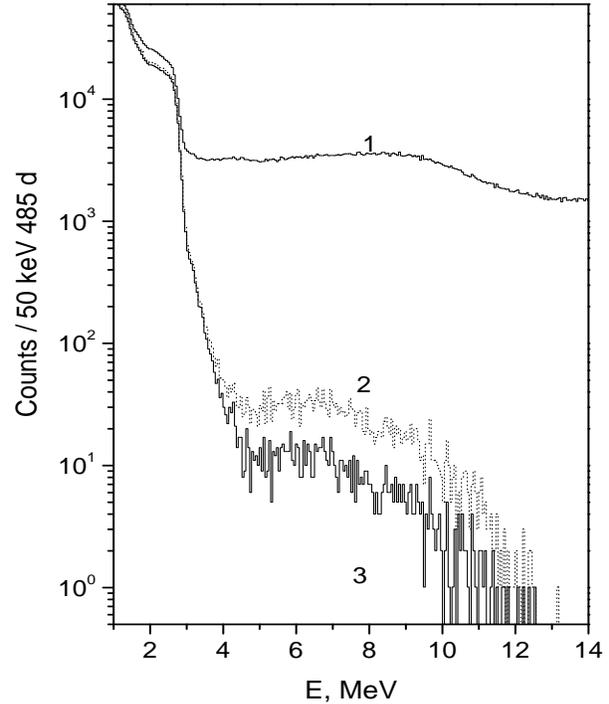


Fig. 4. Energy spectra of the events and effect of the selection cuts. From top to bottom: (1) raw spectrum; (2) with 2 ms muon veto cut; (3) with events within 0.7 s of a muon crossing the SSS removed;

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 reject ^{214}Bi - ^{214}Po coincidences from the ^{238}U chain. Finally, a pulse shape-discrimination analysis based on the Gatti optimal filter [60] is performed to select nucleons. The line (3) shows the events corresponding the positive Gatti variable (see [54] for more details).

4 Results

4.1 Limits on non-Paulian transitions with emission of γ : $^{12}\text{C} \rightarrow ^{12}\tilde{\text{C}} + \gamma$

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

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

$$\tau \geq \varepsilon(\Delta E) \frac{N_N N_n}{S_{lim}} T, \quad (4)$$

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

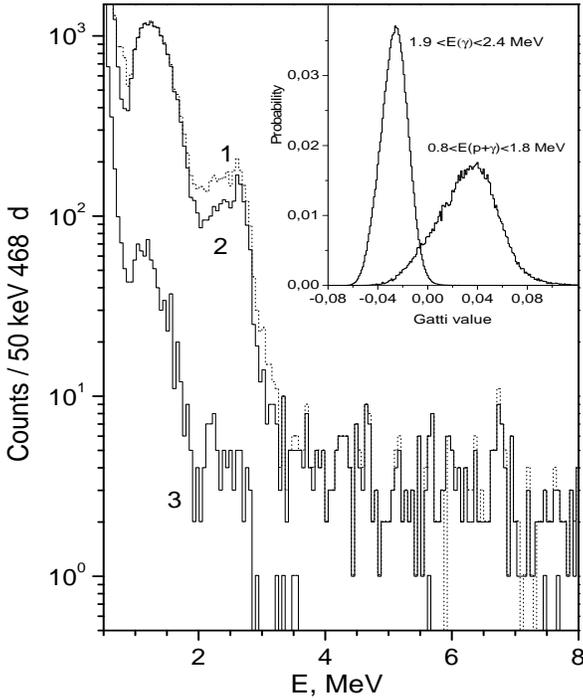


Fig. 5. The energy spectra of events registered inside the FV ($R \leq 3.02$ m). (1) spectrum obtained with 2 ms and 0.7 s muon veto cut; (2) pairs of correlated events (with time interval $\Delta t \leq 2$ ms between signals) are removed; (3) spectrum of the events with positive Gatti variable. In the inset the values of Gatti variable obtained with $^{241}\text{Am}^9\text{Be}$ source for protons and 2.2γ are shown.

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.

As shown in Table 1, 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 ≈ 17.8 MeV. 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 to the γ 's of this energy was found by MC simulations based on GEANT4 code. The uniformly distributed γ 's 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 $N_N = 2.37 \cdot 10^{31}$ (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 $T = 1.282$ y,

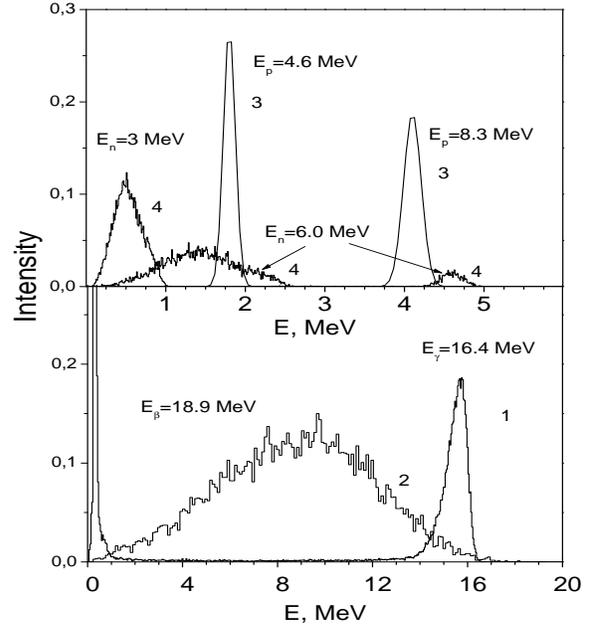


Fig. 6. The response functions of Borexino: 1) $^{12}\text{C} \rightarrow ^{12}\tilde{\text{C}} + \gamma$ (16.4 MeV) decays in IV and 1 m thick layer of buffer; 2) $^{12}\text{C} \rightarrow ^{12}\tilde{\text{N}} + e^- + \bar{\nu}$ (18.9 MeV); 3) $^{12}\text{C} \rightarrow ^{11}\tilde{\text{B}} + p$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

and the upper limit on the number of candidate events is $S_{lim} = 2.44$ with 90% C.L. in accordance with the Feldman-Cousins procedure [61]. The limit obtained using the cited numbers is:

$$\tau_\gamma(^{12}\text{C} \rightarrow ^{12}\tilde{\text{C}} + \gamma) \geq 5.0 \cdot 10^{31} \text{ y}, \quad (5)$$

for the 90% c.l. The result improves by more than 4 orders of magnitude our previous limit, obtained with CTF [50]: $\tau(^{12}\text{C} \rightarrow ^{12}\tilde{\text{C}} + \gamma) \geq 2.1 \cdot 10^{27}$ y. This result is stronger than the one obtained with the NEMO-2 detector $\tau(^{12}\text{C} \rightarrow ^{12}\tilde{\text{C}} + \gamma) \geq 4.2 \cdot 10^{24}$ y [44], and is comparable with the Kamiokande detector for ^{16}O nuclei: $\tau(^{16}\text{O} \rightarrow ^{16}\tilde{\text{O}} + \gamma) \geq 1.0 \cdot 10^{32}$ y for γ 's with energies 19-50 MeV [43]. The limit on the total lifetime of nucleons can be found from the limits on the τ_γ as $\tau = \tau_\gamma \text{Br}(\gamma)$, where $\text{Br}(\gamma) = \Gamma_\gamma / \Gamma_{\text{tot}}$ is the branching fraction of γ -decay. For the case of ^{16}O nucleus the calculated value of $\text{Br}(\gamma)$ is inside interval $(2.7-10.4) \cdot 10^{-5}$ [43]. Unlike the Kamiokande, the Borexino can directly detect the non-Paulian transitions with p -, n - or α -emission.

4.2 Limits on non-Paulian transitions in ^{12}C with proton emission $^{12}\text{C} \rightarrow ^{11}\tilde{\text{B}} + p$.

Using the data of Table 1, one can obtain that the energy released in these transition is within the (5.0-9.0) MeV

interval with a probability of 90%. Taking into account the recoil energy of $^{11}\tilde{\text{B}}$ nucleus, the energy of the proton is (4.6-8.3) MeV.

The response function of protons were simulated by MC code which 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 $^{241}\text{Am}^9\text{Be}$ -source. It was found that the light yield for a proton with the energy $E_p=4.6(8.3)$ MeV corresponds to an electron energy of $E_e=1.8(4.1)$ MeV. It 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 FV cut (line 2, Fig.5). The measured spectrum is fitted by polynomial function and gaussian for proton peak with different positions. Except the region of 2.614 MeV γ peak, this procedure gives $S_{lim} = 52$ at 90% c.l.. The lower limit on the life-time was found from the formula (4) taking into account that $N_N=4.45 \cdot 10^{30}$ for 100 tons FV mass:

$$\tau_p(^{12}\text{C} \rightarrow ^{11}\tilde{\text{B}} + p) \geq 8.9 \cdot 10^{29} \text{ y (90\% c.l.)} \quad (6)$$

The more stringent limit can be obtained by analyzing the spectrum of signals with positive value of Gatti variable (line 3, Fig.5) that corresponds to the detection of α -proton. The lower limit on lifetime is:

$$\tau_p(^{12}\text{C} \rightarrow ^{11}\tilde{\text{B}} + p) \geq 2.1 \cdot 10^{30} \text{ y (90\% c.l.)} \quad (7)$$

where the efficiency of Gatti cut $\varepsilon=0.89$ was taken into account. The corresponding upper limits on nuclear instabilities of ^{12}C nucleus differ from the limits (6) and (7) by factor $N_n=8$.

The obtained upper limits on the nuclear instabilities of ^{12}C are about 4 orders of magnitude stronger than ones obtained with the 300 kg NaI ELEGANT V detector $\tau(^{23}\text{Na}, ^{127}\text{I} \rightarrow ^{22}\tilde{\text{Ne}}, ^{126}\tilde{\text{Te}} + p) \geq 1.7 \cdot 10^{25} \text{ y (90\% c.l.)}$ for protons with $E_p \geq 18$ MeV [45], and with the 250 kg NaI DAMA/LIBRA detector $\tau(^{23}\text{Na}, ^{127}\text{I} \rightarrow ^{22}\tilde{\text{Ne}}, ^{127}\tilde{\text{Te}} + p) \geq 1.9 \cdot 10^{25} \text{ y (90\% c.l.)}$ for protons with $E_p \geq 10$ MeV [35].

The energy of α -particles emitted in $^{12}\text{C} \rightarrow ^8\tilde{\text{Be}} + \alpha$ decay can be found in the 1.0-3.0 MeV interval. Because of the quenching factor, it corresponds to an electron energy range 70-250 keV. The energy 70 keV is close to the Borexino lower energy threshold and 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 $\tau(^{12}\text{C} \rightarrow ^8\tilde{\text{Be}} + \alpha) \geq 6.1 \cdot 10^{23} \text{ y (90\% c.l.)}$.

4.3 Limit on non-Paulian transition in ^{12}C with neutron emission: $^{12}\text{C} \rightarrow ^{11}\tilde{\text{C}} + n$

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 hydrogen-rich media of organic scintillator. The lifetime of neutrons in PC is $\tau \cong 250 \mu\text{s}$, after which they are captured by protons. The cross section for the capture on a proton for a thermal neutron is 0.33 barns. The capture of thermal neutrons $n + p \rightarrow d + \gamma$ is followed by γ - emission with 2.2 MeV energy. The cross sections are much smaller for capture on ^{12}C nuclei ($\sigma_\gamma = 3.5$ mbarns, $E_\gamma = 4.95$ MeV). As a results, the 4.95 MeV peak intensity is about 1% from 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 's with 2.2 MeV energy, and as a result, a limit on the probability of neutron production in the reactions $^{12}\text{C} \rightarrow ^{11}\tilde{\text{C}} + n$. As 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 γ 's of 2.2 MeV energy was precisely measured with $^{241}\text{Am}^9\text{Be}$ neutron source. The position and width of the peak is well known, the fitting procedure gives $S_{lim}=57$. Using equation (4) one can obtain the limit on the probability on neutron emission: $\tau_n(^{12}\text{C} \rightarrow ^{11}\tilde{\text{C}} + n) \geq 8.1 \cdot 10^{29} \text{ y (90\% c.l.)}$

The more stringent limits can be obtained by selecting two consequential events inside the full PC volume - the first signal is from the recoil protons and the second one is 2.2 MeV γ 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 $20 \mu\text{s}$. The energy of the prompt event was set to be $E \geq 0.5$ MeV. The lower threshold is defined by the minimal neutron energy 3.2 MeV (visible energy of 0.6 MeV) taking into the rate of random coincidences. The response functions for neutrons with energy 3.0 and 6.0 MeV are shown in Fig.6. The energy of the second event was required to be $1.0 \text{ MeV} \leq E \leq 2.4 \text{ MeV}$ for detecting the 2.2 MeV γ 's with high efficiency. Additionally, the restored positions of the events have to be within 2 m distance due to the high energy of initial neutron. In such a way, 52 events were selected. Then for different neutron energies E_n inside (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 first excited state of ^{12}C then the high energy part connected with detection of 4.44 MeV γ 's appears in the spectrum of the prompt events. The maximal value of the correlated events $N=26$ was found for the ranges (0.6-2.3) MeV and (4.3-5.0) MeV that corresponds 6 MeV neutrons. Taking into account the probability to find 6.0 MeV neutrons signal in these range ($\varepsilon=0.9$), efficiency of registering 2.2 MeV γ 's ($\varepsilon=0.96$), full number of ^{12}C in the inner vessel $N_N=1.24 \cdot 10^{31}$ and $S_{lim}=33$ for 90% c.l., the limit is:

$$\tau_n(^{12}\text{C} \rightarrow ^{11}\tilde{\text{C}} + n) \geq 3.4 \cdot 10^{30} \text{ y (90\% c.l.)} \quad (8)$$

This result is 8 orders of magnitude stronger than the one obtained through searching for spontaneous neutron

emission from lead: $\tau(\text{Pb} \rightarrow \widetilde{\text{Pb}}+n) \geq 2.1 \cdot 10^{22} \text{ y}$ (68% c.l.) [47].

4.4 Limits on non-Paulian β^\pm -transitions:

$^{12}\text{C} \rightarrow ^{12}\widetilde{\text{N}} + e^- + \bar{\nu}$ and $^{12}\text{C} \rightarrow ^{12}\widetilde{\text{B}} + e^+ + \nu$

The energy released in the reaction $^{12}\text{C} \rightarrow ^{12}\widetilde{\text{N}} + e^- + \bar{\nu}$ is in (16.4 - 21.4) MeV interval. The shape of the β^- spectrum with the most probable end-point energy 18.9 MeV is shown in Fig.6. The spectrum was determined by MC method. The limit on the probability of non-Paulian β^- -transition was based again on the fact of observing no events with $E_e \geq 12.5$ 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}C with respect to the transitions violating the PEP is

$$\tau_{\beta^-} (^{12}\text{C} \rightarrow ^{12}\widetilde{\text{N}} + e^- + \bar{\nu}) \geq 3.1 \cdot 10^{30} \text{ y} \text{ (90\% c.l.)}. \quad (9)$$

This result is 6 orders of magnitude stronger than the one obtained by NEMO-2, $\tau(^{12}\text{C} \rightarrow ^{12}\widetilde{\text{N}} + e^- + \bar{\nu}) \geq 3.1 \cdot 10^{24} \text{ y}$ (90% c.l.) [44].

The data available from the LSD detector [63] situated in the tunnel under Mont Blanc allows obtaining a qualitative limit for this decay mode. In [48], it is claimed that only 2 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}C nuclei. The upper limit that can be obtained using formula (4) with these data (with $S_{lim}=5.91$ events for 90% c.l. and detection efficiency $\varepsilon(E \geq 12 \text{ MeV}) = 0.23$ is $\tau(^{12}\text{C} \rightarrow ^{12}\widetilde{\text{N}} + e^- + \bar{\nu}) \geq 9.5 \cdot 10^{27} \text{ y}$ (90% c.l.).

The end-point energy of the β^+ spectrum is 16.8 MeV, but the spectrum is shifted towards higher energies by $\simeq 0.85$ MeV due to the registering of annihilation quanta (Fig.6). The efficiency of the $^{12}\text{C} \rightarrow ^{12}\widetilde{\text{B}} + e^+ + \nu$ 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}C nuclei is then

$$\tau_{\beta^+} (^{12}\text{C} \rightarrow ^{12}\widetilde{\text{B}} + e^+ + \nu) \geq 2.1 \cdot 10^{30} \text{ y} \text{ (90\% c.l.)} \quad (10)$$

The limits obtained by the NEMO-2 collaboration for this reaction are 6 orders of magnitude weaker: $\tau(^{12}\text{C} \rightarrow ^{12}\widetilde{\text{B}} + e^+ + \nu) \geq 2.6 \cdot 10^{24} \text{ y}$ (90% c.l.) [44].

The final limits on the nucleon instability are shown in Table 2 in comparison with the the previous results obtained for the same PEP-violating transitions. The limit [35] relates to the instability of ^{23}Na and ^{127}I nuclei, all other limits are given per nucleon for which the non-Paulian transition is possible.

4.5 Limits on the relative strength of non-Paulian transitions

The PEP forbidden transitions with emission of γ -, n - or p - and ν , e -pair can be induced by electromagnetic, strong

Table 2. Mean lifetime limits for non-Paulian transitions of nucleons in the Borexino.

Channel	τ_{lim} (y) 90% c.l.	Previous limits
$^{12}\text{C} \rightarrow ^{12}\widetilde{\text{C}} + \gamma$	$5.0 \cdot 10^{31}$	$4.2 \cdot 10^{24}$ (^{12}C) [44] $1.0 \cdot 10^{32}$ (^{16}O) [43]
$^{12}\text{C} \rightarrow ^{11}\widetilde{\text{B}} + p$	$8.9 \cdot 10^{29}$	$1.9 \cdot 10^{25}$ (^{23}Na , ^{127}I) [35]
$^{12}\text{C} \rightarrow ^{11}\widetilde{\text{C}} + n$	$3.4 \cdot 10^{30}$	$2.1 \cdot 10^{22}$ (^{nat}Pb) [47]
$^{12}\text{C} \rightarrow ^{12}\widetilde{\text{N}} + e^- + \bar{\nu}_e$	$3.1 \cdot 10^{30}$	$9.5 \cdot 10^{27}$ (^{12}C) [48, 63]
$^{12}\text{C} \rightarrow ^{12}\widetilde{\text{B}} + e^+ + \nu_e$	$2.1 \cdot 10^{30}$	$2.6 \cdot 10^{24}$ (^{12}C) [44]

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 $\delta^2 = (g_{PV}/g_{NT})^2$ is a measure of the violation of the PEP and represents the mixing probability of non-fermion statistics allowing the transitions to the occupied states. In particular, in quon model of PEP-violation [9,10] the parameter $\delta^2 = \beta^2/2$ corresponds to the probability of 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 P - to S -shell given by the Weisskopf estimate is $\Gamma_\gamma \approx 1.5 \text{ keV}$, then the rate of normal E1 transition is $\lambda = \Gamma_\gamma/\hbar = 2.3 \cdot 10^{18} \text{ s}^{-1}$. With the obtained upper limit on τ_γ (5), the ratio $\delta_\gamma^2 = \widetilde{\lambda} (^{12}\text{C})/\lambda (^{12}\text{C})$ is less than $2.2 \cdot 10^{-57}$ (90% c.l.) This limit is close to result of Kamiokande detector for ^{16}O nuclei - $\delta_\gamma^2 = 2.3 \cdot 10^{-57}$ [43].

Although the E1-transition is the fastest among γ -transitions, the width of hadron emissions is 3-4 orders larger than that of γ -transitions. The widths of single S-hole states in ^{12}C measured for $(p, 2p)$ - and (p, pn) -reaction are $\Gamma_{n,p} \cong 12 \text{ MeV}$ [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 's 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 $\delta_p^2 = \widetilde{\lambda}/\lambda \leq 1.6 \cdot 10^{-59}$ and $\delta_n^2 \leq 4.1 \cdot 10^{-60}$ at 90% c.l.. This result is more then 4 orders of magnitude stronger than the one obtained by DAMA collaboration [35].

The non-Paulian β^\pm -transitions are first-order forbidden $P \rightarrow S$ transitions. The $\log(ft_{1/2})$ values for such first forbidden transitions is 7.5 ± 1.5 . The conservative value $\log(ft_{1/2})=9$ corresponds to life-time $\tau \approx 480 \text{ sec}$ for $Q=18.9 \text{ MeV}$ in the case of β^- -decay (level width is $\Gamma_{\beta^-} \approx 1.4 \cdot 10^{-18} \text{ eV}$) and $\tau \approx 1050 \text{ sec}$ ($Q=17.8 \text{ MeV}$, β^+). As result, the restrictions on the relative strength of non-Paulian β^\pm -decays are significantly lower: $\delta_{\beta^-}^2 \leq 2.1 \cdot 10^{-35}$ and $\delta_{\beta^+}^2 \leq 6.4 \cdot 10^{-35}$ (90% c.l.). The previous

limit $\delta_{\beta^-}^2 \leq 6.5 \cdot 10^{-34}$ obtained in [48] with LSD data [63] is in 30 times weaker. It should be noted, that although the limit for β^\pm -transitions are significantly lower than the nucleon ones, there is a significant difference between these processes. As mentioned above in β^\pm decays new particle (p or n) arises in non-Paulian state, thus Amado-Primakoff arguments may not be valid [13,48]. The limit on $\delta_{\beta^\pm}^2$ can be compared with the limit obtained by the VIP experiment - $\beta^2/2 \leq 4.5 \cdot 10^{-28}$ [38].

The upper limits obtained on the relative strengths of non-Paulian transitions are shown in Table 3. For transitions with (n, p)- and β^\pm -emission the stronger limit is included.

Table 3. Upper limits on the relative strength, $\delta^2 = \tilde{\lambda}/\lambda$ (at 90% C.L.), for non-Paulian transitions in the Borexino.

decay	$\tilde{\lambda}({}^{12}\text{C}),$ (s^{-1})	$\lambda({}^{12}\text{C})$ (s^{-1})	$\delta^2 = \tilde{\lambda}/\lambda$	Previous limits
γ	$5.0 \cdot 10^{-39}$	$2.3 \cdot 10^{18}$	$2.2 \cdot 10^{-57}$	$2.3 \cdot 10^{-57}$ [43]
$N(n, p)$	$7.4 \cdot 10^{-38}$	$1.8 \cdot 10^{22}$	$4.1 \cdot 10^{-60}$	$3.5 \cdot 10^{-55}$ [35]
(e, ν)	$4.1 \cdot 10^{-38}$	$2.0 \cdot 10^{-3}$	$2.1 \cdot 10^{-35}$	$6.5 \cdot 10^{-34}$ [48, 63]

5 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 ${}^{12}\text{C}$ with the emission of γ, n, p and β^\pm particles have been obtained:

$$\begin{aligned} \tau({}^{12}\text{C} \rightarrow {}^{12}\tilde{\text{C}} + \gamma) &\geq 5.0 \cdot 10^{31} \text{ y}, \\ \tau({}^{12}\text{C} \rightarrow {}^{11}\tilde{\text{B}} + p) &\geq 8.9 \cdot 10^{29} \text{ y}, \\ \tau({}^{12}\text{C} \rightarrow {}^{11}\tilde{\text{C}} + n) &\geq 3.4 \cdot 10^{30} \text{ y}, \\ \tau({}^{12}\text{C} \rightarrow {}^{12}\tilde{\text{N}} + e^- + \nu) &\geq 3.1 \cdot 10^{30} \text{ y} \end{aligned}$$

and

$$\tau({}^{12}\text{C} \rightarrow {}^{12}\tilde{\text{B}} + e^+ + \bar{\nu}) \geq 2.1 \cdot 10^{30} \text{ y},$$

all with 90% C.L.

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

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