УДК 539.184

# NEW EXPERIMENTAL LIMITS ON THE ELECTRON STABILITY AND EXCITATION OF NUCLEAR LEVELS IN <sup>23</sup>Na, <sup>127</sup>I AND <sup>129</sup>Xe INDUCED BY THE ELECTRON DECAY ON THE ATOMIC SHELL

P. Belli<sup>a</sup>, R. Bernabei<sup>a</sup>, C. J. Dai<sup>b</sup>, H. L. He<sup>b</sup>, G. Ignesti<sup>c</sup>, A. Incicchitti<sup>c</sup>, H. H. Kuang<sup>b</sup>, J. M. Ma<sup>b</sup>, F. Montecchia<sup>a</sup>, O. A. Ponkratenko<sup>d</sup>, D. Prosperi<sup>c</sup>, V. I. Tretyak<sup>d</sup>, Yu. G. Zdesenko<sup>d</sup>

<sup>a</sup>Dip. di Fisica, Universita' di Roma «Tor Vergata» and INFN, sez. Roma2, I-00133 Rome, Italy <sup>b</sup> IHEP, Chinese Academy, P.O. Box 918/3, Beijing 100039, China

<sup>c</sup> Dip. di Fisica, Universita' di Roma «La Sapienza» and INFN, sez. Roma, I-00185 Rome, Italy
<sup>d</sup> Institute for Nuclear Research, MSP 03680 Kiev, Ukraine

The background measurements have been performed in the Gran Sasso National Laboratory of INFN with the help of the large mass highly radiopure  $\simeq 100 \text{ kg}$  DAMA NaI(Tl) set-up (34866 kg · day statistics) and the  $\simeq 6.5 \text{ kg}$  liquid Xenon DAMA scintillator set-up (2257.7 kg · day). New life time limits on the charge nonconserving (CNC) electron decays have been established:  $\tau(e^- \rightarrow \nu_e \overline{\nu}_e \nu_e) > 4.2(2.4) \cdot 10^{24} \text{ y}$  and  $\tau(e^- \rightarrow \nu_e \gamma) > 3.4(2.0) \cdot 10^{26} \text{ y}$  at 68 % (90 %) C.L. Life time limits on the CNC electron capture with nuclear levels excitation of  $^{23}$ Na,  $^{127}$ I, and  $^{129}$ Xe are also established; they are in the range  $\tau > 1.5 \cdot 10^{23} - 4 \cdot 10^{24} \text{ y}$ . All limits are at least few times higher than the ones previously available.

В подземной лаборатории Гран-Сассо были проведены измерения фона с использованием установок DAMA NaI(Tl) с массой  $\simeq 100$  кг (статистика 34866 кг · сут) и 6,5 кг жидкого ксенона (2257,7 кг · сут). Установлены новые экспериментальные пределы на время жизни электрона:  $\tau(e^- \rightarrow \nu_e \overline{\nu}_e \nu_e) > 4,2(2,4) \cdot 10^{24}$  лет и  $\tau(e^- \rightarrow \nu_e \gamma) > 3,4(2,0) \cdot 10^{26}$  лет на 68 % (90 %) уровне достоверности. Установлены пределы на время жизни электрона с возбуждением ядерных уровней в  $^{23}$ Na,  $^{127}$ I и  $^{129}$ Xe:  $\tau > 1,5 \cdot 10^{23} - 4 \cdot 10^{24}$ лет. Все полученные пределы в несколько раз выше установленных ранее.

#### **INTRODUCTION**

Since electron is the lightest electrically charged particle, the stability of the electron implies the conservation of electric charge. In the framework of the standard quantum electrodynamics, the charge conservation is a direct consequence (Weinberg theorem [1]) of massless photons which are imposed by the fundamental underlying principle of gauge invariance. Nevertheless, the possibility that the electric charge conservation may be broken in future unified gauge theories and the related implications have been intensively discussed in literature [2–7]. Although no self-consistent theory describing electric charge nonconservation has been yet constructed (see for details reviews [6] and refs. therein), many efforts have been devoted to test this fundamental feature of the nature in direct experiments [8–19] since the early search by Feinberg and Goldhaber in 1959 [8].

The idea of the pioneering experiment [8] was to use a NaI(Tl) scintillator and to look for the X ray and Auger electrons cascade, which would follow the decay of a K electron in a Iodine atom (energy release is 33.2 keV). This approach — named «disappearance» approach — is sensitive to all the decay modes giving decay particles which escape the detector without depositing energy (for example:  $e^- \rightarrow \nu_e \overline{\nu}_e \nu_e$ ). Another approach, sensitive to the  $e^- \rightarrow \nu_e \gamma$  decay mode, searches for 255.5 keV  $\gamma$  quantum; in this case electron decays in the surrounding materials will contribute as well as the ones inside the detector. All the results available in literature — for both types of experiments — are summarized in Table 1. The best limits on the mean life of the electron previously established were: in the «disappearance» channel  $\tau_e > 1.3 \cdot 10^{24}$  y [19] and for  $e^- \rightarrow \nu_e \gamma$  decay  $\tau_e > 3.7(2.1) \cdot 10^{25}$  y at 68 % (90 %) C.L. [17]<sup>1</sup>.

Detector	Volume, cm <sup>-3</sup>	Limit on	Limit on	Year [ref.]
		$\tau_e(e^- \rightarrow \nu_e \bar{\nu}_e \nu_e), y$	$\tau_e(e^- \rightarrow \nu_e \gamma), y$	
NaI(Tl)	1287	$1.0\cdot10^{18}$	$1.0\cdot 10^{19}$	1959 [8]
NaI(Tl)	348	$2.0\cdot10^{21}$	$4.0 \cdot 10^{22}$	1965 [9]
Ge(Li)	66	$5.3 \cdot 10^{21}$	—	1975 [10]
NaI(Tl)	1539	$2.0\cdot 10^{22}$	$3.5 \cdot 10^{23}$	1979 [11]
Ge(Li)	130	$2.0\cdot 10^{22}$	$3.0 \cdot 10^{23}$	1983 [12]
Hp-Ge	135	—	$1.5(1.1) \cdot 10^{25}$	1986 [13]
Hp-Ge	3 · 140	$2.7(1.7) \cdot 10^{23}$	_	1991 [14]
NaI(Tl)	$17 \cdot 10570$	$1.2\cdot10^{23}$	—	1992 [15]
Hp-Ge	591	—	$2.4(1.2) \cdot 10^{25}$	1993 [16]
Hp-Ge	$48 + 2 \cdot 209$	$4.3(2.6) \cdot 10^{23}$	$3.7(2.1) \cdot 10^{25}$	1995 [17]
LXe	2000	$1.5 \cdot 10^{23}$	$2.0(1.0) \cdot 10^{25}$	1996 [18]
Hp-Ge	132	$1.3 \cdot 10^{24}$	—	1998 [19]
NaI(Tl)	9 · 2643	$4.2(2.4) \cdot 10^{24}$	—	1999 [20] & this work
LXe	2000		$3.4(2.0) \cdot 10^{26}$	2000 [21] & this work

Table 1. Experimental limits on the electron life time at 68 % (90 %) C.L.

Another method of searching for the disappearance of electrons on atomic shells involving nuclear levels excitation was proposed and realized at first by Holjevic et al. [23]. The idea is to consider the possible influence of the electron decay on the atomic nucleus. The exploited process, analogous to an electron capture, does not change the nucleon charge but leaves the nucleus in an excited state:  $(A, Z) + e^- \rightarrow (A, Z)^* + \nu_e$ . Possible mechanisms of such CNC processes were considered in Refs. 23, 24, where their advantages for the CNC quest, involving the CNC nuclear excitation through both the weak boson and photon mediating

<sup>&</sup>lt;sup>1</sup>It has been shown that the decay channel  $e^- \rightarrow \nu_e \gamma$  may be affected — in the case of explicit violation of the electric charge conservation due to the presence in the Lagrangian of a CNC term — by the catastrophic emission of a huge amount of longitudinal bremsstrahlung photons with very small energies; thus the decay of an electron will not be accompanied by the 255.5 keV  $\gamma$  rays [6]. On the contrary, the filling of the shell after the electron disappearance would occur before the emission of soft photons and will not be affected by them (see [17] and refs. therein). Therefore the «disappearance»  $\tau$  limit is considered more «safe» and model independent, as credited by the Particle Data Group [22].

processes, have been pointed out. The CNC electron capture can feed the excited states of the nucleus with energies  $E_{\rm exc}$  up to  $m_e c^2 - E_B$  ( $E_B$  is the binding energy of the electron). In the de-excitation process the nucleus returns to the ground state emitting one or more  $\gamma$  quanta and conversion electrons which could be observed by a suitable detector. It is supposed that CNC excitations feed preferably the lowest levels with difference in spin between ground and excited states  $\Delta J = 0, 1$  and that K electrons are most probably involved in the process, being the closest to the nucleus. Results of previous experiments to search for the CNC electron capture are summarized in Table 2.

Nucleus,	Efficiency	Excluded area	Lifetime limits $\tau$ , y			
$E_{\rm exc}$	$\eta$	of effect $S$	[25,26] & this work	[23]	[24]	
		90 % C.L.	90% C.L.	90 % C.L.	68 % C.L.	
<sup>23</sup> Na						
440.0 keV	0.60	3009	$1.5 \cdot 10^{23}$			
$^{127}$ I						
57.6 keV	0.99	3149	$2.4 \cdot 10^{23}$	$2.1 \cdot 10^{21}$	$5.8 \cdot 10^{22}$	
202.9 keV	0.89	3418	$2.0 \cdot 10^{23}$	$1.9  \cdot  10^{21}$	$5.6 \cdot 10^{22}$	
375.0 keV	0.72	3061	$1.8 \cdot 10^{23}$	$2.4 \cdot 10^{21}$		
418.0 keV	0.61	2974	$1.6 \cdot 10^{23}$	$2.4 \cdot 10^{21}$		
<sup>129</sup> Xe						
39.6 keV	0.99	18.5	$1.1 \cdot 10^{24}$			
236.1 keV	0.97	5.5	$3.7 \cdot 10^{24}$			
318.2 keV	0.65	6.1	$2.2 \cdot 10^{24}$			
321.7 keV	0.67	5.6	$2.5 \cdot 10^{24}$			
411.5 keV	0.50	4.6	$2.3 \cdot 10^{24}$			

*Table 2.* Experimental lifetime limits on the CNC electron capture involving nuclear levels excitation of <sup>23</sup>Na, <sup>127</sup>I, and <sup>129</sup>Xe

This paper describes the new improved limits on the electron instabilities which were obtained as a «by-product» results of the DAMA data taking with two low-background detectors operating in the Gran Sasso National Laboratory of INFN: DAMA NaI(Tl) scintillator with mass near 100 kg [27, 28] and 6.5 kg DAMA liquid Xe (LXe) scintillator enriched in <sup>129</sup>Xe at 99.5 % [18, 29], mainly dedicated to the particle Dark Matter direct search. The results of present work were previously published in Refs. 20, 21, 25, 26 where more details on measurements and data processing can be found.

# 1. EXPERIMENTAL SET-UPS AND MEASUREMENTS

**1.1. Measurements with NaI(Tl) Detectors.** The detailed description of the highly radiopure  $\simeq 100$  kg DAMA set-up and its performances are discussed in Ref. 28. Here we briefly recall the main features of this apparatus.

The data were collected with nine 9.70 kg NaI(Tl) crystal scintillators enclosed in radiopure Cu housings. Each detector has two 10 cm long tetrasil-B light guides directly coupled to

the opposite sides of the bare crystal. Two photomultipliers (PMT) EMI9265-B53/FL work in coincidence and collect light at single photoelectron threshold, while 2 keV is the software energy threshold [27, 28]. The detectors are enclosed in a low radioactive Cu box inside a low radioactive shield made of 10 cm Cu and 15 cm Pb; the last is surrounded by 1.5 mm Cd foils and about 10 cm of polyethylene. A high purity (HP) Nitrogen atmosphere is maintained inside the Cu box by a continuous flux of HP Nitrogen gas from bottles stored underground for a long time. The whole shield is wrapped in Supronyl and maintained also in the HP Nitrogen atmosphere. The installation is subjected to air conditioning (the maximum level of temperature variation is  $< 0.2 \,^{\circ}$ C) to avoid any influence of the temperature on the light yield of the crystals, on the PMT's spectral sensitivity and gain and on the stability of the electronics. This allows one to keep constant the energy scale, the energy resolution and the energy threshold of the detectors, as verified also by the continuous monitoring of the stability parameters and by the routine energy scale calibrations. A pulse shape analysis was used to reject the residual noise by exploiting the different time structure of the PMT noise (fast pulses with decay time of order of tens ns) and scintillation signals (decay time of order of hundreds ns).

In the calibration measurements the typical energy resolution is  $\sigma/E = 7.5 \%$  at 59.5 keV. The knowledge of the energy scale was assured by periodical calibration with <sup>241</sup>Am source and by monitoring the position and resolution of the <sup>210</sup>Pb peak (46.5 keV) present at level of few cpd  $\cdot$ kg<sup>-1</sup> in the measured energy distributions. This peak is mainly due to a surface contamination by environmental Rn occurred during the first period of the crystals storage deep underground. The standard deviation of the position of this peak estimated for all nine detectors during about 180 days — without applying any correction — does not exceed 1.2 % [27,28]; thus the effect is negligible considering the continuous calibration monitoring and correction. In conclusion, owing to the mentioned procedures, the energy scale, the energy resolution and the energy threshold of the detectors are well established.

**1.2. Measurements with Liquid Xenon Detector.** The LXe DAMA set-up ( $\simeq 6.5$  kg — i. e.,  $\simeq 21$  — of liquid Xenon scintillator) and its performance has been published in Refs. 29 and only the main features of the detector are described here.

The gas used is Kr-free Xenon enriched in <sup>129</sup>Xe at 99.5%. The U/Th contamination of <sup>129</sup>Xe does not exceed  $\approx 2$  ppt at 90% C.L. The vessel for the LXe is made of OFHC low radioactivity copper ( $\leq 100 \ \mu Bq \cdot kg^{-1}$  for U/Th and  $\leq 310 \ \mu Bq \cdot kg^{-1}$  for K). The scintillation light collection is assured by three EMI PMTs with MgF<sub>2</sub> windows, working in coincidence. Their measured quantum efficiency — for normal incidence — ranges between 18 and 32% at the LXe scintillation wavelength (175 nm) with a flat behaviour around this value. The PMTs collect the scintillation light through three windows (3" in diameter) made of special cultured crystal quartz (total transmission of the LXe ultraviolet scintillation light is  $\approx 80$ %, including the reflection losses). A low radioactivity Cu shield inside the thermo-insulation vacuum cell surrounds the PMTs; then, 2 cm of steel (insulation vessel thickness), 5–10 cm of low radioactivity Cu, 15 cm of low radioactivity Pb,  $\approx 1$  mm of Cd and  $\approx 10$  cm of polyethylene are used as outer hard shielding. The environmental Rn near the external insulation vessel of the detector is removed by continuously flushing high purity (HP) Nitrogen gas (from bottles stored underground for a long time) inside a sealed Supronyl envelope, which wraps the whole shield.

Each PMT is connected with a low noise preamplifier. For every event the following data are stored: (i) amplitudes of each PMT pulse and (ii) amplitude and shape of the sum pulse

(recorded by a Lecroy transient digitizer). The energy dependence of the detector resolution was measured [29] and can be expressed as following:  $\sigma/E = 0.056 + 1.19/\sqrt{E}$ , where  $\sigma$  is in keV. Some other information can be found in Ref. 29.

# 2. DATA ANALYSIS AND DISCUSSION OF THE RESULTS

**2.1. Decay**  $e^- \rightarrow \nu_e \overline{\nu}_e \nu_e$ . The idea of the present work is to use the distinguished features of the DAMA NaI(Tl) set-up to look for signals from X ray and Auger electron cascade, which would follow the decay not only of a K electron (energy released 33.2 keV) but also of a L electron (energy release of about 5 keV) in a Iodine atom.

Each Iodine atom contains 8 electrons on L shell (two electrons on L1, two on L2, and four on L3 subshell), while only 2 are available on K shell. Thus, the possibility to investigate the energy region corresponding to L-shell electron decays will increase the source strength by a factor 4 with respect to the standard procedure searching for K-electron decay. The study of the L-electron decay is possible here owing to the low energy threshold and the low background rate of the DAMA set-up.

The statistics considered in the present analysis is 19511 kg day (DAMA/NaI-1 & 2 running periods) [27]. The 2–20 keV energy distributions of each detector can be found in Refs. 27. Since the behaviours of these distributions in the energy region of interest here are not very different, the cumulative energy distribution (Fig. 1) can be used for the electron lifetime estimate.



Fig. 1. Cumulative experimental energy distribution measured by all the detectors in the region of interest for the process searched for; the statistics is 19511 kg  $\cdot$  day. The dotted line represents the result of a fit given by the sum of a linear function (simplified background model suitable for the present purposes) and of the sum of the three gaussians associated to the process searched for; this last contribution requires only one free parameter (see text)

The possible decay of L electrons in Iodine atoms inside the NaI(Tl) detectors would be visible as a peak at the energy of about 5 keV (5.19 keV for L1 shell, 4.85 keV for L2 shell and 4.56 keV for L3 shell [30]) with  $\sigma/E$  corresponding to the detector energy resolution. The absence of such a peak in the collected data is evident in Fig. 1. Thus, the experimental spectrum can be used to determine the upper limit of the electron life time using the formula:  $\tau = (\eta Nt)/S$ , where  $\eta$  is the detection efficiency; N is the number of electrons on L shell

of Iodine atoms; t is the measuring time and S is the number of events due to the effect searched for and excluded with given C.L.

The cascade of low energy X rays and Auger electrons with the same energy of about 5 keV will be absorbed in a large NaI(Tl) crystal giving an efficiency  $\eta = 1$ . Nine 9.70 kg detectors include  $3.51 \cdot 10^{26}$  NaI molecules, that correspond to  $2.81 \cdot 10^{27}$  electrons on L shell of Iodine atoms. Thus, the total Nt is equal to  $1.72 \cdot 10^{27}$  electrons  $\cdot$ y. As the simplest estimate of the excluded number of events S we can accept the standard statistical deviation of the total number of events in the 3.5-6.0 keV energy region. The latter is a very sensitive interval which offers a practically symmetric window centered around the centroid of the 3 peaks and including 66% ( $\eta_{\text{window}}$ ) of the total area. The value  $S = (\delta w)/\eta_{\text{window}} = 482(793)$  with 68% (90%) C.L. is found; there  $\delta$  (0.0165 cpd  $\cdot$ kg<sup>-1</sup>) is the standard deviation of the total rate in the 3.5-6.0 keV energy interval and w is the statistics (19511 kg  $\cdot$ day). The obtained result gives the following limit of the electron lifetime:  $\tau(e^- \rightarrow \nu_e \overline{\nu}_e \nu_e) > 3.6(2.2) \cdot 10^{24}$  y with 68% (90%) C.L.

Then, with the aim to make the estimation of S more accurate, the experimental energy distribution in the interval 3.5–6.0 keV was fitted by the sum of two functions: the background and the effect being searched for. As simplified background model, suitable for the present purposes, the linear function has been assumed there. The effect has been represented by the sum of three gaussians, centred at 4.56, 4.85, and 5.19 keV respectively, and with energy resolutions scaled here according to:  $\sigma/E \propto 1/\sqrt{E}$ . The amplitudes of the gaussians have been normalized for two electrons on L1, two electrons on L2, and four electrons on L3shell (requiring, therefore, only one free parameter for the effect amplitude). From the fit the amplitude of the effect was found to be  $(-0.0029 \pm 0.0240)$  cpd  $\cdot$  kg<sup>-1</sup>, giving no statistical evidence for it ( $\chi^2$ /d.o.f. was 1.2). Using these values the upper limit on the events number S was calculated according to the Particle Data Group procedure [31]. In fact, from the amplitude of the effect given by the fit, the lower limit 0.02118 (0.03663) cpd  $\cdot$  kg<sup>-1</sup> at 68 % (90%) C.L. can be estimated, giving: S < 413(715) and  $\tau(e^- \rightarrow \nu_e \overline{\nu}_e \nu_e) > 4.2(2.4) \cdot 10^{24}$  y at 68 % (90 %) C.L. This result is near 3 times higher than the best limit previously established in the experiment with HP-Ge detectors, where the «disappearance» of Ge K-shell electrons was studied [19].

The searches for «disappearance» of electrons on the atomic shells are also related with the experimental quest for the violation of the Pauli exclusion principle (PEP). The transition of electrons to fully filled L shell — process usually forbidden by PEP — will result in an energy release equal to the binding energy of electron on L-shell. From an experimental point of view, both processes are undistinguishable in NaI(Tl) detector; thus the established limit on  $\tau_e$  could be regarded also as a limit on the probability of the PEP violation.

**2.2.** Decay  $e^- \rightarrow \nu_e \gamma$ . The idea of the present work is to search for  $\gamma$  rays (with the energy of  $\approx 255$  keV) which could accompany the possible decay of any electron in the LXe scintillator (and in its surroundings) by analyzing the energy distribution collected during about 347 days. The exact value of the total energy deposited in the detector depends on the place where the electron decay occurs. If it happened outside the detector,  $\gamma$  quantum with initial energy  $E_{\gamma} = (m_e c^2 - E_B)/2$  can hit the detector and release some energy in it ( $E_B$  is the binding energy of the electron on the corresponding atomic shell). If electron decays inside the detector, the additional energy release from X rays and Auger electrons following the atomic de-excitation (with the total energy of  $E_B$ ) should be also taken into account.

#### 64 Belli P. et al.

The experimental spectrum of the LXe scintillator in the energy region 40-500 keV with total statistics of 2257.7 kg  $\cdot$  day is shown in Fig. 2, where the absence of the peak searched for around  $\approx 255$  keV is evident. Therefore these data can be used to set the bound on the probability of the electron decay in the considered channel.



Fig. 2. Energy spectrum measured by the LXe scintillator in the energy region 40–500 keV (total statistics: 2257.7 kg · day). In the inset the part of the spectrum in the energy region of interest is shown in linear scale together with the fitting curve (dashed line) and excluded peak ( $\tau = 2.0 \cdot 10^{26}$  y)

To estimate the lifetime limit  $\tau$ , we use the formula:  $\tau = t \sum_i (\eta_i N_i) / S$ , where  $\eta_i$  is the efficiency to detect  $\approx 255$  keV  $\gamma$  quanta from the *i*th medium (Xe detector and Cu vessel; consecutive layers of steel, Cu and Pb shield were found to be negligible);  $N_i$  is the number of atomic electrons in the corresponding *i*th medium; *t* is the measuring time; and S is the number of the effect's events, which can be excluded with the given confidence level on the basis of the experimental data. The  $\eta_i$  values and the response functions of the LXe scintillator were evaluated through the Monte Carlo simulation with the help of GEANT3.21 package [32]. The Doppler broadening of the measured  $\gamma$  lines due to the electrons' movement in different atomic shells was also taken into account [16]. Doppler-broadened line shape is represented by the sum of 17 Gaussians for the case of Xe atoms and of 10 Gaussians for Cu atoms; however, considering the energy resolution of the LXe scintillator, its total response function to the effect searched for is very close to a Gaussian centred at  $\approx 255$  keV with FWHM equal to 80 keV. The calculated values of the detection efficiency are  $\eta_{\rm Xe} = 85 \%$  and  $\eta_{\rm Cu} = 10 \%$ .

The S value has been determined in two ways.

First, it has been evaluated by using the so-called «one  $\sigma$  approach» in which the excluded number of the effect's events is estimated simply as the square root of the number of background counts in a suitably chosen energy window  $\Delta E$ . Notwithstanding its simplicity, this method gives the right scale of the sensitivity of the experiment. For instance, 34 counts are present within the interval 205-325 keV which contains 90% of the expected peak; thus, the square-root estimate gives S < 6.5events. Using this S value, the numbers of electrons in the LXe detector and Cu vessel  $(N_{\rm Xe} = 1.64 \cdot 10^{27}; N_{\rm Cu} = 4.67 \cdot 10^{27})$ , the measuring time and the calculated efficiencies, we obtain the limit  $\tau > 2.7 \cdot 10^{26}$  y. Furthermore, the S value was determined by using the standard least squares procedure, fitting the experimental energy distribution in the neighborhood of the peak searched for by the sum of a simplified background model (exponent) and of the effect's peak (represented by the simulated response function of the detector as described above). From the fit, the peak's area equal to  $-2.4 \pm 6.3$  counts  $(\chi^2/d.o.f. = 0.67)$  has been obtained, giving no evidence for the effect. Then, the number of the effect's events, which can be excluded at 90% (68%) C.L. is calculated [22] as 9.0(5.2), giving the limit  $\tau > 2.0(3.4) \cdot 10^{26}$  y at 90% (68%) C.L. The fitting curve and the excluded peak are shown in the inset of Fig.2. The present bound on the  $e^- \rightarrow \nu_e \gamma$  decay channel is one order of magnitude higher than the best limit  $(2.1 \cdot 10^{25} \text{ y at } 90 \% \text{ C.L.})$  previously obtained for this decay mode [17].

In accordance with the results of Ref. 33, the transition probability for the electron de-cay  $e^$ form:  $(m_e/m_{\gamma})^2 \simeq \varepsilon_{e\nu\gamma}^2 (\alpha/32\pi)(m_ec^2/\hbar)(m_e/m_{\gamma})^2,$ where  $\alpha$  is the fine structure constant, and the  $\varepsilon_{e\nu\gamma}$  gives a measure of the charge nonconservation. From this equation we can find numerically  $\varepsilon_{e\nu\gamma}^2 (m_e/m_{\gamma})^2 < 5.6 \cdot 10^{-25}/\tau_{e\nu\gamma} (\tau_{e\nu\gamma})^2$ is in years), thus our experimental limit leads to the bound  $\varepsilon_{e\nu\gamma}^2 (m_e/m_{\gamma})^2 < 2.8(1.6) \cdot 10^{-51}$  at 90 % (68 %) C.L. The latter expression, combined with the best laboratory limit on the photon mass  $m_{\gamma} < 2 \cdot 10^{-16}$  eV [34] yields the restrictions  $\varepsilon_{e\nu\gamma}^{2'} < 4.3(2.5) \cdot 10^{-94}$  at 90 % (68 %) C.L.

From the other hand, we can use the established  $\tau_{e\nu\gamma}$  limit to find the bound on the photon mass. The relation between  $\tau_{e\nu\gamma}$  and  $m_{\gamma}$  was found, for example in the framework of the SU(5) model [35] as following:  $\tau_{e\nu\gamma} \simeq 10^{-25} (m_Z/m_{\gamma})^2$  y, where  $m_Z = 91.2$  GeV is the mass of the Z bozon. Using this relation and our value  $\tau_{e\nu\gamma} > 3.4 \cdot 10^{26}$  y, we can receive  $m_{\gamma} < 1.6 \cdot 10^{-15}$  eV.



Fig. 3. Energy spectrum measured by the LXe scintillator in the energy region 40–500 keV with total statistics of 823.1 kg · day. In the inset the low energy part is shown in linear scale together with the fitting curve and excluded peak ( $\tau =$ 1.1·10<sup>24</sup> y) for the first excited level of <sup>129</sup>Xe

2.3. Nuclear Levels Excitation Due to the Electron Decay. The data accumulated with the DAMA NaI(Tl) and LXe detectors were used also to establish the limits on the probability of the process in which an electron disappears from the atomic shell and the nucleus is left in an excited state. Such process is analogous to the usual electron capture but does not change the nucleus' charge:  $(A, Z) + e^- \rightarrow (A, Z)^* + \nu_e$ .

The CNC electron capture can feed the excited states of the nucleus with energies  $E_{\rm exc}$ up to  $m_ec^2 - E_B$  ( $E_B$  is the binding energy of the electron in the considered atomic shell). It is supposed that CNC excitation feeds preferably the lowest levels with difference in spin between ground and excited state  $\Delta J = 0, 1$ , and that K electrons (being the closest to the nucleus) most probably are involved in the process. In the de-excitation the nucleus returns to the ground state emitting  $\gamma$  quanta and conversion electrons. X rays and Auger electrons emitted in the relaxation of the atomic shell should be also taken into account.

The results of the previous experiments [23, 24], performed with NaI(Tl) detectors, are given in Table 2. In this paper the first investigation of the CNC electron capture involving nuclear levels excitation of  $^{129}$ Xe is presented; the statistics considered here is 823.1 kg · day [26].

Five levels of <sup>129</sup>Xe could be excited due to the studied process with  $E_{\text{exc}} = 39.6; 236.1;$ 318.2; 321.7 and 411.5 keV [30]. Taking into account the binding energy of the Xe K atomic shell ( $E_B^K = 34.6$  keV), the energies of the possible peaks in the background spectra should be: 74.2; 236.1<sup>1</sup>; 352.8, 356.3, and 446.1 keV. The experimental spectrum of the LXe scintillator in the energy region 40-500 keV is shown in Fig. 3, where the absence of these peaks is evident. Thus limits can be set for the probabilities of CNC nuclear excitations of <sup>129</sup>Xe nuclei. We estimate the life time limits  $\tau$  using the formula  $\tau = (\eta N t)/S$ , where  $\eta$ is the detection efficiency, N is the number of electrons on K shell of Xe atoms; t is the measuring time, and S is the number of the effect's events, which can be excluded with a given C.L. To calculate the  $\eta$  values, de-excitation processes in <sup>129</sup>Xe nuclei inside the LXe scintillator and the response function of the detector were simulated with the help of GEANT3.21 package [32]; the code DECAY4 [36] was used for description of the initial event's kinematics. Calculated efficiencies are varied from 0.99 for  $E_{\rm exc} = 39.6$  keV to 0.50 for  $E_{\rm exc} = 411.5$  keV (Table 2). The S values were determined in two ways. Firstly, by using the so-called «one  $\sigma$  approach», in which the excluded number of the effect's events is estimated simply as square root of the number of background counts in a suitably chosen energy window  $\Delta E$ . For instance, in the measured spectrum within the energy interval 45– 103 keV (it contains 95% of expected 74.2 keV peak area) there are 129 counts; thus, the square root estimate gives S < 11.4 events. Using this value S, total number of K electrons in the LXe detector ( $N = 6.0 \cdot 10^{25}$ ), measuring time and calculated efficiency, we obtain the limit  $\tau > 1.7 \cdot 10^{24}$  y (68 % C.L.) for the 74.2 keV peak. The results for other peaks are within  $\tau > (3-7) \cdot 10^{24}$  y at 68 % C.L. Further, S values were determined by using the standard least squares procedure, where the experimental energy distribution in the neighborhood of the peak searched for was fitted by the sum of background (exponential behaviour for the first peak and a straight line for the others) and signals peak being sought. As the last one the response function of the detector was simulated by a gaussian with the proper width. For example, the obtained area for the first peak (74.2 keV) is  $-11 \pm 15$  counts ( $\chi^2$ /d.o.f. value is 1.3), thus giving no evidence for the signal. Then, the number of the signal events, which can be excluded with 90 % (68 %) C.L. were calculated [22] as 18.5 (10.4). It gives the limit  $\tau > 1.1(2.0) \cdot 10^{24}$  y at 90% (68%) C.L. for the first ( $E_{\rm exc} = 39.6$  keV) excited level of <sup>129</sup>Xe. The excluded number of signal events for other levels obtained by a similar procedure and the corresponding  $\tau$  limits are shown in Table 2. For illustration the fitting curve and excluded peak for the first excited level is depicted in the inset of Fig. 3.

The data accumulated with the DAMA NaI(Tl) detectors were also analyzed to set the limits on the CNC nuclear excitations in <sup>23</sup>Na and <sup>127</sup>I nuclei; the statistics used was 34866 kg  $\cdot$  day [25]. The obtained results are summarized in Table 2. Other details can be found in Ref. 25.

Possible mechanisms of the CNC electron capture were discussed in [23–26]. Such process could include both the weak boson and photon exchange. Comparing the CNC electron capture with the standard electron capture and with the standard internal conversion process and using the obtained lifetime limits, the bounds on the CNC admixtures in the weak interactions were found:  $\varepsilon_W^2 < 2.2 \cdot 10^{-26}$  and  $\varepsilon_\gamma^2 < 1.3 \cdot 10^{-42}$  at 90% C.L. The detailed discussion is given in Refs. 25, 26.

<sup>&</sup>lt;sup>1</sup>Because the second excited level is long-lived ( $t_{1/2} = 8.89$  d), the energy of peak searched for is equal to  $E_{\text{exc}} = 236.1$  keV.

### CONCLUSION

Using the the low energy threshold and the low background rate of the  $\approx 100$  kg DAMA NaI(Tl) set-up, the electron stability has been studied by looking for the signal from X ray and Auger electron cascade which would follow the decay («disappearance») of any L electron of one of the Iodine atoms. The obtained lifetime limit  $\tau(e^- \rightarrow \nu_e \overline{\nu}_e \nu_e) > 4.2(2.4) \cdot 10^{24}$  y at 68 % (90 %) C.L. is few times higher than the best limit previously achieved by considering the K-shell electrons in Ge detectors.

The highest limit on the lifetime of the electron for the decay  $e^- \rightarrow \nu_e \gamma$  was established with the super-low background DAMA set-up with the liquid Xe scintillator:  $\tau(e^- \rightarrow \nu_e \gamma) > 3.4(2.0) \cdot 10^{26}$  y at 68 % (90 %) C.L.

Lifetime limits on the CNC electron capture with nuclear levels excitation of <sup>23</sup>Na, <sup>127</sup>I, and <sup>129</sup>Xe were obtained; they are in the range  $\tau > 1.5 \cdot 10^{23} - 4 \cdot 10^{24}$  y. All limits are at least few times higher than the ones previously available.

The severe restrictions on the CNC admixtures in the weak interactions and on the photon mass were derived:  $\varepsilon_W^2 < 2.2 \cdot 10^{-26}$ ,  $\varepsilon_\gamma^2 < 1.3 \cdot 10^{-42}$ ,  $\varepsilon_{e3\nu}^2 < 1.1 \cdot 10^{-26}$ ,  $\varepsilon_{e\nu\gamma}^2 < 2.5 \cdot 10^{-94}$ , and  $m_\gamma < 1.6 \cdot 10^{-15}$  eV [20,21,25,26].

#### REFERENCES

- 1. Weinberg S. // Phys. Rev. B. 1964. V. 135. P. 1049.
- 2. Okun L. B., Zeldovich Ya. B. // Phys. Lett. B. 1978. V. 78. P. 597.
- 3. Voloshin M. B., Okun L. B. // JETP Lett. 1978. V. 32. P. 145.
- 4. Mohapatra R. N. // Phys. Rev. Lett. 1987. V. 59. P. 1510.
- 5. Okun L. B. Leptons and Quarks. Amsterdam, 1982. P. 181.
- Okun L. B. // Sov. Phys. Usp. 1989. V. 32. P. 543; Comments Nucl. Part. Phys. 1989. V. 19. P. 99; Phys. Rev. D. 1992. V. 45, No. 11. P. VI.10.
- 7. Rubakov V.A. // Part. and Nuclei, Letters. 2001. No. 3[106]. P. 106.
- 8. Feinberg G., Goldhaber M. // Proc. Nat. Acad. Sci. USA. 1959. V.45. P. 1301.
- 9. Moe M. K., Reines F. // Phys. Rev. B. 1965. V. 140. P. 992.
- 10. Steinberg R. I. et al. // Phys. Rev. D. 1975. V. 12. P. 2582.
- 11. Koval'chuk E. L. et al. // JETP Lett. 1979. V. 29. P. 145.
- 12. Bellotti E. et al. // Phys. Lett. B. 1983. V. 142. P. 435.
- 13. Avignone F. T. III et al. // Phys. Rev. D. 1986. V. 34. P. 97.
- 14. Reusser D. et al. // Phys. Lett. B. 1991. V. 255. P. 143.
- 15. Ejiri H. et al. // Phys. Lett. B. 1992. V. 282. P. 281.
- 16. Balysh A. et al. // Phys. Lett. B. 1993. V. 298. P. 278.

68 Belli P. et al.

- 17. Aharonov Y. et al. // Phys. Lett. B. 1995. V. 353. P. 168; Phys. Rev. D. 1995. V. 52. P. 3785.
- 18. Belli P. et al. // Astropart. Phys. 1996. V. 5. P. 217.
- 19. Klimenko A. A. et al. // Yad. Fiz. 1998. V. 61. P. 1229 (in Russian).
- 20. Belli P. et al. // Phys. Lett. B. 1999. V. 460. P. 236.
- 21. Belli P. et al. // Phys. Rev. D. 2000. V. 61. P. 117301.
- 22. Particle Data Group. Review of Particle Physics // Phys. Rev. D. 1996. V. 54. P. 1.
- 23. Holjevic S. et al. // Phys. Rev. C. 1987. V. 35. P. 341.
- 24. Ejiri H. et al. // Phys. Rev. C. 1991. V. 44. P. 502.
- 25. Belli P. et al. // Phys. Rev. C. 1999. V. 60. P. 065501.
- 26. Belli P. et al. // Phys. Lett. B. 1999. V. 465. P. 315.
- Bernabei R. et al. // Phys. Lett. B. 1996. V. 389. P. 757; 1997. V. 408. P. 439; 1998. V. 424. P. 195; 1999. V. 450. P. 448; 2000. V. 480. P. 23; Nuovo Cim. A. 1999. V. 112. P. 1541; Belli P. et al. // Phys. Rev. D. 1999. V. 61. P. 023512.
- 28. Bernabei R. et al. // Nuovo Cim. A. 1999. V. 112. P. 545.
- 29. Belli P. et al. // Nuovo Cim. C. 1996. V. 19. P. 537; Phys. Lett. B. 1996. V. 387. P. 222; Bernabei R. et al. // Phys. Lett. B. 1998. V. 436. P. 379.
- 30. Firestone R. B. Table of Isotopes. 8th ed. N.Y.: John Wiley & Sons, 1996.
- 31. Feldman G. J., Cousins R. D. // Phys. Rev. D. 1998. V. 57. P. 3873.
- 32. GEANT. CERN Program Library Long Write-up W5013. CERN, 1994.
- 33. Suzuki M. // Phys. Rev. D. 1988. V. 38. P. 1544.
- 34. Lakes R. // Phys. Rev. Lett. 1998. V. 80. P. 1826.
- 35. Huang J. C. // J. Phys. G. 1987. V. 13. P. 273.
- 36. Ponkratenko O.A. et al. // Phys. At. Nucl. 2000. V. 63. P. 1282.