Abstract  New lifetime limits on the charge non-conserving (CNC) electron capture with excitation of the 417.9 keV nuclear level in the $^{127}$I are established by using the coincidence technique. The analysed exposure is 0.87 ton $\times$ yr, collected deep underground at the Gran Sasso National Laboratory of the INFN by the highly radiopure DAMA/LIBRA setup ($\approx$250 kg of highly radiopure NaI(Tl)). The new limit on the mean life is $\tau > 1.2 \times 10^{24}$ yr (90 % C.L.), about one order of magnitude larger than those previously available for CNC electron capture involving nuclear level excitations of $^{127}$I and of the same order of magnitude than those achieved for analogous processes in $^{129}$Xe.

1 Introduction

The conservation of the electric charge is one of the fundamental laws of standard quantum electrodynamics based on the underlying principle of gauge invariance. The invariance of the QED Lagrangian under U(1) local gauge transformations requires massless photons and exact charge conservation (CC) in accordance with the Weinberg theorem [1]. Nevertheless, the possibility that charge conservation may be broken in future unified gauge theories and the implications of such a violation have been extensively discussed in the literature [2–8]. The symmetry breaking can be either explicit or spontaneous, and—in the latter case—it can be triggered by a suitable charged Higgs field. Moreover, modern extra-dimension theories (as e.g. the brane-world scenario [9, 10]) offer new challenges for the searches of charge non-conserving (CNC) processes and, in particular, for the disappearance of charged particles. In fact, in those scenarios the charge is conserved in the multi-dimensional space, while the CC violation is due just to the observational limits. The theoretical estimates of the lifetimes expected for similar processes depend on the number of extra-dimensions; considering electrons disappearance, the expected lifetime for 3 extra-dimensions is $9 \times 10^{25}$ yr [11], that is not far from the present experimental sensitivities [12].

The experimental efforts to test this basic feature of the nature in direct measurements have been continuing since the pioneering search by Feinberg and Goldhaber [13]. The electric charge conservation can be tested for electrons and nucleons separately (see reviews [6–8] and references therein). For instance, the highest lifetime limit published for the electron’s disappearance (by looking for X-ray and Auger electron cascades which would follow the decay of an atomic electron) is in NaI(Tl) detectors $\tau_e > 2.4 \times 10^{24}$ yr at 90 % C.L. [12]. For the particular decay mode $e^- \rightarrow v_e \gamma$ (whose signature is the presence of a 255.5 keV $\gamma$ ray) the highest published limit is $\tau > 6.6 \times 10^{26}$ yr at 90 % C.L. in PXE detector [14]; another recent result in HPGe is given in [15]. It is worth noting that, in case of explicit violation of the electric charge conservation due to the presence in the Lagrangian of a $L_{\text{CNC}}$ term, this latter decay channel may be affected by the catastrophic emission of a huge amount of longitudinal bremsstrahlung photons with very small energies, and the decay of an electron will not be accompanied by the 255.5 keV $\gamma$ ray [6–8]. On the contrary, the filling of the atomic shell after the electron disappearance would occur before the emission of soft photons and will not be af-
fected by them (see Ref. [16, 17] for discussions and references). Therefore, at present the disappearance lifetime limit is considered more reliable and model independent [18].

The possible CNC processes involving simultaneously both electrons and nucleons were less extensively searched for and some experimental data are available. For the particular case of the CNC-β decay, the studied nuclei were: \(^{87}\text{Rb} \rightarrow ^{87m}\text{Sr}\) [19–21] (the best lifetime limit is \(\tau > 7.5 \times 10^{19}\) yr at 90 % C.L. [21]); \(^{113}\text{Cd} \rightarrow ^{113m}\text{In}\) (\(\tau > 1.4 \times 10^{18}\) yr at 90 % C.L.) [22]; \(^{71}\text{Ga} \rightarrow ^{71}\text{Ge}\) (\(\tau > 2.3 \times 10^{23}\) yr at 90 % C.L. [23] and \(\tau > 3.5 \times 10^{26}\) yr at 68 % C.L. [24], respectively); \(^{73}\text{Ge} \rightarrow ^{73}\text{As}\) (\(\tau > 2.6 \times 10^{23}\) yr [25] at 90 % C.L.); \(^{136}\text{Xe} \rightarrow ^{136}\text{Cs}\) (\(\tau > 1.3 \times 10^{23}\) yr at 90 % C.L. [26]); \(^{115}\text{In} \rightarrow ^{115m}\text{Sn}\) (\(\tau > 4.1 \times 10^{20}\) yr at 90 % C.L. [27]); \(^{139}\text{La} \rightarrow ^{139}\text{Ce}\) [28] (\(\tau > 1.0 \times 10^{18}\) yr at 90 % C.L.); \(^{100}\text{Mo} \rightarrow ^{100}\text{Tc}\) (\(\tau > 4.5 \times 10^{19}\) yr at 90 % C.L. [29]).

In 1987 Holjevic et al. [30] were the first to propose and exploit the search for the electron’s disappearance in atomic shells involving the excitation of nuclear levels. The idea was to consider the process, analogous to the electron capture (EC), which does not change the nucleon charge but leaves the nucleus in an excited state: \((A, Z) + e^- \rightarrow (A, Z)^* + \nu_e\). If an electron disappears from an atomic shell, the energy \(E = m_e c^2 - E_B\) is available for the nucleus excitation and for the neutrino kinetic energy; \(E_B\) is the binding energy of the electron. Possible mechanisms of such CNC processes were considered in Refs. [30, 31] and the advantages of their investigation for the CNC search were also pointed out. The CNC nuclear excitation involves both the weak boson and the photon-mediating processes [31]; moreover, while the electron decay is concerned with the CNC process at the lepton sector, the nuclear excitation (and the nuclear β decay) is concerned with the CNC process at both the lepton and quark (nucleon) sectors. Currently there is no self-consistent and non-contradictory theory, describing the possible violations of the charge conservation and allowing to derive restrictions on theoretical parameters from experimental lifetime limits on CNC processes. Bahcall has attempted to explain charge non-conservation in terms of weak interactions [32, 33] including a small CNC part that has the usual form except for a neutrino replacing the electron in the lepton current. In this point of view one can write \(|M_{\text{CNC}}|^2 \approx \varepsilon^2 |M_{\text{CC}}|^2\) where \(M_{\text{CNC}}\) is the matrix element associated with CNC and \(M_{\text{CC}}\) is the conventional matrix element for a process in which charge is conserved. In Refs. [30, 31] two CNC processes (both called CNC electron capture) have been considered using this approach, their diagrams are presented in Fig. 1. In Fig. 1(a) charge conservation is violated in the hadron sector; in Fig. 1(b) CC is violated in the lepton sector. The analogous CC processes are shown in Fig. 1(c) electron capture and Fig. 1(d) internal electron conversion.

The CNC electron capture can feed the excited states of the nucleus with energies \(E_{\text{exc}}\) up to \(m_e c^2 - E_B\). In the deexcitation process the nucleus returns to the ground state emitting \(\gamma\) quanta and conversion electrons which could be observed by a suitable detector. If the electron capture takes place in the detector itself, the observed energies will be shifted up to the \(E_{\text{exc}} + E_B\) value due to the absorption of X-rays and Auger electrons emitted in the relaxation of the atomic shells. The emission of \(\gamma\) quanta in coincidence with atomic relaxation gives a clear signature for this process in a suitable experimental setup, where the search for these events in coincidence allows the reduction of the background and the improvement of the experimental sensitivity.

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.

In the first experiment [30] an NaI(Tl) scintillator (\(\approx 7500\) cm\(^3\) volume) was used to search for the possible excitation of \(^{127}\text{I}\) nuclear levels by looking for the deexcitation \(\gamma\) rays. The obtained limit for the CNC electron capture process was \(\tau > 2 \times 10^{21}\) yr at 90 % C.L. [30] for all the excited levels. This result was followed by the one of the Oskawa group (\(\tau > 6 \times 10^{22}\) yr at 68 % C.L. measured only for the first \(57.6\) keV and the second \(202.8\) keV excited states), which used an array of NaI(Tl) detectors in the Kamioka underground laboratory [31]. Afterwards, DAMA/LXe experimental setup investigated the possible CNC-EC decays [34] with the production of \(^{129}\text{Xe}\) in an excited state; the best obtained limit on this process is \(\tau > 3.7 \times 10^{24}\) yr at 90 % C.L. The results obtained with the DAMA/NaI setup were published in Ref. [35] for all the excited states obtaining \(\tau > 2.4 \times 10^{23}\) yr at 90 % C.L. During the last
years the large mass highly radiopure NaI(Tl) setup developed by the DAMA Collaboration [36] was successfully applied to rare event searches [37], mainly to the dark matter particles investigation [38, 39]. The unique features of the DAMA/LIBRA apparatus [36] are here used to study the CNC electron capture involving nuclear level excitation of $^{127}$I by means of the coincidences technique.

2 Experimental set-up

The results presented in the following have been obtained by analysing the data collected by the DAMA/LIBRA (Large sodium Iodide Bulk for Rare processes) set-up [36–39], whose description, radiopurity and main features are discussed in details in the dedicated Ref. [36]. The sensitive part of this set-up is made of 25 highly radiopure NaI(Tl) crystal scintillators (5-rows by 5-columns matrix); each NaI(Tl) detector has 9.70 kg mass and a size of $(10.2 \times 10.2 \times 25.4) \text{ cm}^3$. The bare crystals are enveloped in Tetratec-teflon foils and encapsulated in radiopure OFHC Cu housing. In each detector two 10 cm long special quartz light guides act also as optical windows on the two end faces of the crystal and are coupled to two low background photomultipliers (built by Electron Tubes Limited) working in coincidence at single photoelectron threshold. The light response is typically 5.5–7.5 ph.e./keV depending on the detector. The detectors are housed in a sealed low-radioactive copper box installed in the center of a low-radioactive Cu/Pb/Cd-foils/polyethylene/paraffin shield; moreover, about 1 m concrete (made from the Gran Sasso rock material) almost fully surrounds (mostly outside the barrack) this passive shield, acting as a further neutron moderator. The copper box is maintained in HP Nitrogen atmosphere in slightly overpressure with respect to the external environment; it is part of the threefold-level sealing system which excludes the detectors from environmental air. A hardware/software system is operative to monitor the running conditions, and self-controlled computer processes automatically control several parameters and manage alarms. For the radiopurity, the electronic chain, the data acquisition system and for all the other details see Refs. [36, 39].

3 Experimental results

The usual approach to investigate CNC electron capture processes is to search in the experimental energy distribution for the peaks due to the $\gamma$ rays induced by possible deexcitation processes in the $^{127}$I nuclei which might follow the CNC electron capture.

In accordance with the low-energy level schemes of $^{127}$I (Fig. 2) four levels could be excited in $^{127}$I ($E_{\text{exc}} = 57.6; 202.8; 375.0$ and 417.9 keV). Thus, taking into account the binding energies of iodine K atomic shells ($E_{K}^B = 33.2$ keV), possible peaks to be searched for in the experimental energy distribution, when all the energy is detected (with the source = detector approach), are at: 90.8, 236.0, 408.2, and 451.1 keV.

An alternative experimental approach to study the CNC electron capture is instead considered in the present work. The idea is that each CNC electron capture decay produces an excited level and the relaxation of the atomic shells. In a multi-detector setup the products of the atomic relaxation are contained in the source = detector releasing a total energy of 33.2 keV, considering only $^{127}$I K-shell electrons involved in the process. The $\gamma$ quanta emitted in the nucleus deexcitation may escape from the source = detector and then interact with one of the surrounding detectors, giving events in coincidence with multiplicity two (two detectors fire). The search for events with multiplicity two in the particular energy interval offers both a peculiar signature for this process and a significant reduction of the background.

A Montecarlo simulation (based on EGSnrc code [40]) has been realized to study the possibility to search in DAMA/LIBRA for coincidences from the deexcitation and the relaxation of the atomic shells. Although in the scenario of Refs. [30, 31] the most probable process involves the 57.6 keV excited level, the present analysis is focused on the case of the 417.9 keV excited level of $^{127}$I, since it offers the largest efficiency for the detection of double-coincidence from the CNC electron capture searched for, and the lowest background.

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Figure 3-left shows for this process the scatter plot of the energies released in two detectors for simulated events with multiplicity two; there are two clear spots due to the correlation of the events with energy 33.2 keV in one detector with those with energy 417.9 keV in the other detector. In Fig. 3-right the events collected by DAMA/LIBRA with multiplicity two in the particular energy interval show the coincidence signature of the CNC electron capture searched for, giving events in coincidence with multiplicity two (two detectors fire). The search for events with multiplicity two in the particular energy interval offers both a peculiar signature for this process and a significant reduction of the background.
ity 2 are shown; for comparison the same number of events fills the two scatter plots. As it can be seen, in the experimental scatter plot there is no evidence of these spots. This leads to the conclusion that there is no evidence in the experimental data for the process searched for; therefore, just a lower limit on the lifetime of this CNC-EC process can be set.

Figure 4-left shows—for the process searched for—the simulated energy distribution of events in one detector in case of coincidence with multiplicity 2 and selecting the energy of the event in the other detector in the 27–40 keV window (that is, about $2\sigma$ around 33.2 keV considering the DAMA/LIBRA energy resolution), where the atomic relaxation products can give signal. Figure 4-right shows the experimental energy distribution collected with the same criteria; again there is no evidence of the effect searched for. Thus, only a lower limit on the lifetime of the process searched for can be derived by using the formula: 

$$
\tau = \frac{\eta N_0 t}{S_{\text{lim}}},
$$

where $\eta$ is the detection efficiency, $N_0$ is the number of electrons on the K shell of the iodine atoms, $t$ is the measuring time and $S_{\text{lim}}$ is the number of events of the effect searched for, which can be excluded at a given confidence level on the basis of the experimental data.

To have a preliminary determination of the experimental sensitivity for the lifetime of the CNC-EC decay with production of the excited level at 417.9 keV of $^{127}$I the so called 1$\sigma$-approach can be used; there the excluded number of the events due to the effect searched for is estimated simply as the square root of the number of background counts in the chosen energy window. In the energy distribution of

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**Fig. 3** Scatter plot of events with multiplicity 2: first event’s energy versus the second event’s energy. **Left**: Montecarlo expectation for the CNC-EC decay in the 417.9 keV excited level. **Right**: experimental data. For comparison the same number of events fills the two scatter plots.

**Fig. 4** **Left**: Expected energy distribution of the CNC electron capture events with multiplicity 2 and with the condition that the energy of the events in the other detector is in the 27–40 keV window, calculated by Montecarlo code considering the excitation of the 417.9 keV level of $^{127}$I. See text. **Right**: Experimental energy distribution around 417.9 keV of events with multiplicity 2 and with the condition that the energy of the events in the other detector is in the 27–40 keV window. The signal of the deexcitation $\gamma$ quanta following the CNC-EC process is expected to be in this window (see text). The superimposed red line shows the best-fit curve ($\chi^2/d.o.f. = 1.04$) (Color figure online)
the coincidence events (see Fig. 4-right) where the energies of the first and of the second detector in the pair are in the 27–40 keV and in the 372–464 keV energy windows ($\pm 2\sigma$ around the expected peak’s energy 417.9 keV), respectively, one gets the upper limit: $S_{\text{lim}} = 140$ events (68 % C.L.). Then considering the number of K electrons in the iodine atomic shell in 1 kg of NaI(Tl) ($8.03 \times 10^{24}$ kg$^{-1}$), the measuring time (0.87 ton $\times$ yr of exposure) and the $\eta$ efficiency (averaged over all the pairs of detectors as estimated by the Monte Carlo code; for double coincidences with 27–40 keV and 372–464 keV energy windows: $\eta = 0.041$), the CNC-EC lifetime lower limit: $\tau > 2.0 \times 10^{24}$ yr (68 % C.L.), is derived.

A refined analysis has been performed by fitting the experimental data with a function sum of an exponential function (to describe the background contribution) and a gaussian (describing the peak searched for, centered at 417.9 keV and with energy resolution $\sigma \sim 23$ keV, see Ref. [36]). For the CNC-EC signal the obtained value is $S = -(260 \pm 296)$ events ($\chi^2/d.o.f. = 1.04$). Thus, the number of events searched for is compatible with zero and using the Feldman and Cousins procedure [41] the upper limit on the number of events due to CNC-EC process can be written as: $S_{\text{lim}} = 264$ events (90 % C.L.), corresponding to the lower limit on the lifetime (in this case $\eta = 0.045$): $\tau > 1.2 \times 10^{24}$ yr (90 % C.L.). For the sake of completeness, we note that no sizeable displacement from the previous outcome is obtained if adopting in the fitting procedures other reasonable functions to describe the background behavior in the energy interval of interest; for example, if a straight line function is used, the value $S = -(244 \pm 304)$ events ($\chi^2/d.o.f. = 0.95$) is derived, giving rise to a limit on the lifetime well compatible with the one quoted above.

Following the Bahcall approach the obtained lifetime limit can be used to estimate the CC violation. Using the procedure of Ref. [35], the $\epsilon^2$ in case of CNC-EC mediated by W-boson or by $\gamma$-exchange has been determined (for the theoretical parameters we have used here those tabulated in Refs. [42, 43]): $\epsilon_W^2 < 2.5 \times 10^{-25}$ and $\epsilon_{\gamma}^2 < 5.0 \times 10^{-39}$. It is worth nothing that the $\epsilon^2$ estimated for the CNC-EC processes depends on the considered excited level. Since the lowest energetic level is favoured, in Ref. [35] the limits on $\epsilon^2$—calculated on the lowest lying level of $^{127}$I—are more stringent, although the sensitivity in $\tau$ is lower than that achieved in the present work. However, in absence of a rigorous fundamental theory of CC violation there is no way to know “a priori” if CC breaking gives CNC-EC via either W or $\gamma$ exchange or mainly producing an excited level or another one. The $\epsilon^2$ parameter, in fact, is dependent on the used modeling. The missing of a self-consistent model for CNC processes and considering that for fundamental questions—like the one under discussion—any “a priori” argument based either on pure aesthetic or on other principles could give wrong results (as it was demonstrated, for instance, with parity conservation); at some extent we could face unexpected things. These arguments lead to conclude that the approach considered to derive $\epsilon^2$ can give a preliminary idea on the reached sensitivity for CC violation, but on the other hand can give wrong results in absence of a rigorous theory for CNC processes. The lifetime determination on the contrary has an absolute value for each possible (past, present or future) theory and it is not model dependent.

### 4 Conclusions

Possible CNC-EC process with production of the excited level 417.9 keV of $^{127}$I has been investigated by using the coincidence technique; thanks to this approach the background for this process has been reduced by a significant factor. The obtained limit on this CNC-EC process is $\tau > 1.2 \times 10^{24}$ yr (90 % C.L.); it improves of about one order of magnitude the best one previously available [35]. The obtained limit is also very close to the best limit available in literature for CNC-EC processes $\tau > 3.7 \times 10^{24}$ yr (90 % C.L.) in $^{129}$Xe [34].

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