Physics Letters B 711 (2012) 41–45

Contents lists available at SciVerse ScienceDirect

Physics Letters B

www.elsevier.com/locate/physletb

Search for ^{7}Li solar axions using resonant absorption in LiF crystal: Final results

P. Belli [a], R. Bernabei [a,b,*], F. Cappella [c,d], R. Cerulli [e], F.A. Danevich [f], A. d'Angelo [c,d], A. Incicchitti [c,d], V.V. Kobychev [f], M. Laubenstein [e], O.G. Polischuk [f], V.I. Tretyak [f]

[a] INFN, Sezione di Roma "Tor Vergata", I-00133 Rome, Italy
[b] Dipartimento di Fisica, Università di Roma "Tor Vergata", I-00133 Rome, Italy
[c] INFN, Sezione di Roma "La Sapienza", I-00185, Rome, Italy
[d] Dipartimento di Fisica, Università di Roma "La Sapienza", I-00185, Rome, Italy
[e] INFN, Laboratori Nazionali del Gran Sasso, I-67010 Assergi (AQ), Italy
[f] Institute for Nuclear Research, MSP 03680 Kyiv, Ukraine

ARTICLE INFO

Article history:
Received 27 January 2012
Received in revised form 22 March 2012
Accepted 25 March 2012
Available online 27 March 2012
Editor: V. Metag

Keywords:
Axions
HP Ge gamma detector
Resonant excitation

ABSTRACT

The resonance excitation of the ^{7}Li nuclei in a LiF crystal with mass of 553 g by hypothetical axions emitted in the deexcitation of the ^{7}Li nuclei in the Sun was searched for deep underground at the Gran Sasso National Laboratories (LNGS) of INFN (3600 m w.e.). The data collected with a low background HP Ge detector 244 cm^3 during 4044h have allowed us to set the limit on the axion mass: $m_a < 8.6$ keV (90% C.L.) which is the best one for the ^{7}Li solar axions to-date.

1. Introduction

The axion (a) is a hypothetical particle arising in an elegant solution of the so-called "strong CP problem" of quantum chromodynamics (QCD) proposed by Peccei and Quinn [1]. As in the weak interactions, the CP symmetry is not supposed to be conserved in the strong interactions too. The general form of the QCD Hamiltonian contains a term with amplitude θ which violates the CP symmetry [2]. In particular, this leads to the neutron electric dipole moment d_n, which, however, is not experimentally observed; only a strong limit is known as $d_n < 2.9 \times 10^{-26}$ e cm [3]. This transforms to limit $\theta < 10^{-10}$ instead of naturally expected value of $\theta \sim 1$. It should be also noted that θ not only is small, but in fact it is the difference between two big quantities of different origin that makes the situation even more unnatural [2].

In 1977 Peccei and Quinn [1] introduced a new global (PQ) symmetry which is spontaneously violated at some energy scale $E \simeq f_a$, that totally suppresses the θ term. Weinberg [4] and Wilczek [5] found that the PQ symmetry leads to the existence of a new pseudo-scalar neutral particle (named the axion) with spin 0 and mass $m_a \simeq 6 \times 10^6/f_a$ (m_a in eV, f_a in GeV). The interaction of the axion with different components of usual matter is characterized by different coupling constants: $g_{a\gamma}$ (interaction with photons), g_{ae} (with electrons), g_{aN} (with nucleons); they also are proportional to $1/f_a$. In the very beginning, it was supposed that for the "standard" (PQWW, following the names of the authors) axion $f_a \simeq 250$ GeV and $m_a \simeq 100$ keV. However, very soon axions with such masses were experimentally excluded [2,6–8]. Other models were proposed which supposed f_a values up to 10^{19} GeV: KSVZ (Kim [9] and Shifman, Vainstein and Zakharov [10]), and DFSZ (Dine, Fischler and Srednicki [11] and Zhitnitskii [12]). In these models the axion mass could be very small: $m_a \simeq 10^{-12}$ eV. Because also $g_{ai} \sim 1/f_a$ are very small, such axions often are named invisible.

If axions exist, they could be intensively born inside the Sun. They could be produced: (1) in the interaction of the thermal γ quanta with the solar electromagnetic fields due to the Primakoff effect; the energy spectrum of these axions is continuous up to ~ 20 keV with the mean value of ~ 4.2 keV. Their flux is related with value of coupling constant $g_{a\gamma}$; (2) axions can be emitted instead of γ quanta in deexcitations of excited nuclear levels in magnetic transitions; their spectrum is quasi-monoenergetic, and the flux is related with g_{aN}. Nuclei can be excited due to the thermal movement (in the center of the Sun the temperature is $\simeq 1.3$ keV); evidently, nuclei with excited levels at low energies E_{exc} are preferable (^{57}Fe: $E_{exc} = 14.4$ keV, ^{83}Kr: $E_{exc} = 9.4$ keV). Also, the nuclear levels can be populated in the nuclear reactions

---

[*] Corresponding author at: Dip. di Fisica, Università di Roma "Tor Vergata", I-00133 Rome, Italy.
E-mail address: rita.bernabei@roma2.infn.it (R. Bernabei).

0370-2693/$ – see front matter © 2012 Elsevier B.V. All rights reserved.
http://dx.doi.org/10.1016/j.physletb.2012.03.067

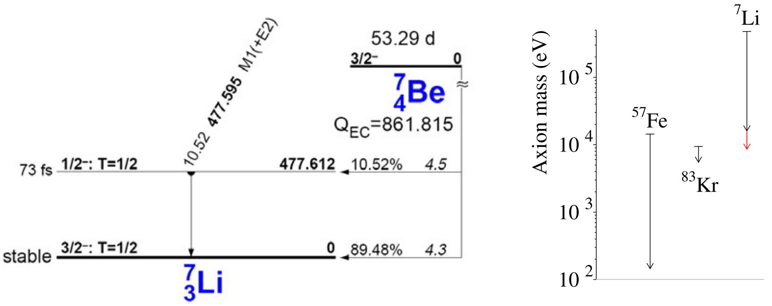

Fig. 1. (Color online.) Left: Decay scheme of ^{7}Be in accordance with [13]. Right: Current status of m_a limits obtained in searches for quasi-monoenergetic solar axions through resonant excitation of nuclei. For ^{7}Li, the situation is shown before (in black) and after (in red) the efforts described in [14] and in this work (see discussion in the text).

inside the Sun, e.g. in the main *pp* cycle: $^7\text{Be} + e^- \to {}^7\text{Li} + \nu_e$ with 10.52% population of the ^{7}Li level with 477.6 keV energy. The decay scheme of the ^{7}Be in terrestrial conditions is shown in Fig. 1 (left).

In spite of a great theoretical interest in axions (which also could constitute a part of dark matter, see e.g. [15]), intensive experimental searches up to now gave no direct evidences of their existence. Indirect cosmological and astrophysical arguments prefer the axion mass in the range of $10^{-6} - 10^{-2}$ eV [2,8,16,17]. The laboratory searches for axions are based on several possible mechanisms of the axion interactions with the ordinary matter: (1) conversion of axion to photon in a laboratory magnetic field or in a crystal detector (inverse Primakoff effect); (2) Compton conversion of axion to photon $a + e \to \gamma + e$ (analog of the Compton effect); (3) decay of axion to two photons $a \to \gamma\gamma$; (4) axioelectric effect of interaction with an atom $a + (A, Z) \to e + (A, Z)$ (analog of photoelectric effect); (5) resonant absorption of axions emitted in nuclear magnetic transitions in some radioactive source (unstable isotope, nuclear reactor or the Sun) by the analogue nuclei in a target. Because these mechanisms are based on different kinds of interaction of the axion with matter, they are sensitive to different coupling constants ($g_{a\gamma}$, g_{ae}, g_{aN}), and diverse experiments are mutually complementary [3]. While in most of the experiments the axion–photon coupling constant $g_{a\gamma}$ is involved (or combination of $g_{a\gamma}$ and g_{aN}, etc.), in the mechanism (5) the probability of axion emission at birth and of its capture is only related with the coupling constant of axions with the nucleons g_{aN}; the uncertainties related with $g_{a\gamma}$, g_{ae} disappear. Further details can be found in [2,3,6–8,16,17].

The resonant excitation of nuclei as an experimental scheme of searching for quasi-monoenergetic solar axions was proposed in [18]. Quasi-monoenergetic axions emitted in magnetic transitions instead of γ quanta by excited nuclei (^{7}Li, ^{57}Fe, ^{83}Kr or others) in the Sun could resonantly excite corresponding levels of the same nuclei on the Earth. ^{7}Li, as a favorable target to search for hadronic solar axions, was discussed at the first time in [19]. Due to the motion of ^{7}Li nuclei in the Sun core with high temperature (near 1.3 keV in the center), the axion 478 keV line is symmetrically Doppler broadened with width $\simeq 0.5$ keV that is much bigger than the energy of the nuclear recoil ($\simeq 1.8 \times 10^{-2}$ keV), than the redshift due to the gravitation of the Sun ($\simeq 5 \times 10^{-3}$ keV), and than the decay width of the 478 keV excited level ($\simeq 6 \times 10^{-6}$ keV). Because of the thermal broadening, the 478 keV axions could be resonantly absorbed by ^{7}Li nuclei in a laboratory at the Earth [19].

Gamma quanta (and/or conversion electrons) emitted in the subsequent deexcitation processes can be observed with the help of detectors located near a sample with ^{7}Li, ^{57}Fe, ^{83}Kr (or incorporating these nuclei). The positive effect was not observed to-date in any performed experiment, and only limits on the probability of the process (and corresponding values of the coupling constant g_{aN} and axion mass m_a) were obtained. The summary of all the experiments devoted to the search for the resonant excitation of nuclei by quasi-monoenergetic solar axions is given in Table 1. All the experiments were performed at the Earth level, except [14] and this work which were carried out underground (LNGS, 3600 m w.e.). Current status of m_a limits obtained in this experimental approach is presented in Fig. 1 (right) where for each nucleus an arrow (started at the corresponding value of E_{exc} and finished at the limit's value) shows the excluded m_a values. It should be noted that searches for ^{7}Li axions have some advantages because their flux is directly related with the main *pp* cycle and thus with the luminosity of the Sun, in contrast to axions related with ^{57}Fe and ^{83}Kr due to some uncertainties in their solar abundances and in the distribution of the temperature inside the Sun.

As one can see from Table 1, the best m_a limits in the experiments looking for the resonant excitation of nuclei by monoenergetic solar axions were obtained in measurements with ^{57}Fe. However, it should be noted that, because the energy of ^{57}Fe excited level is 14.4 keV, axions with mass greater than 14.4 keV (if they exist) just cannot be emitted instead of γ quanta in ^{57}Fe deexcitation (the same concerns also ^{83}Kr). This is the reason why experiments with ^{7}Li, which has greater excitation energy: 477.6 keV, are also valuable. It is evident also that it is important to set in ^{7}Li measurements m_a limit lower than 14.4 keV; this extends a window in excluded axion masses to limits of [477.6, 0.145] keV.

In 2008, we performed preliminary studies [14] of a few LiF samples (LiF powders and LiF crystals) to investigate the possibilities to improve the best ^{7}Li limit: $m_a < 16.0$ keV, which existed at that time [20]. Low background measurements of LiF powder (with mass of 243 g) performed underground during 722h at LNGS allowed us to set the limit $m_a < 13.9$ keV, in spite of a significant pollution of the powder by U/Th natural radioactive chains [14]. The measurements also showed that the LiF crystals, in contrast with the LiF powders, are radioactively pure (only limits on U/Th pollutions were set), and it was evident that further measurements should be done with this material. Here we report the final results of the experiment performed with a radiopure LiF crystal (553 g) during 4044h of measurements.

2. Experimental measurements

The data were collected with a LiF crystal[1] ⌀89 × 40 mm having a mass of 552.6 g and with an ultra-low background HP Ge detector (GeBer, 244 cm^3, with carbon fibre window 0.76 mm thick) installed deep underground (3600 m w.e.) at the Gran Sasso

---

[1] Produced by the Czochralski method in the Institute for Scintillation Materials (Kharkiv, Ukraine). The crystal was doped by W on the level of $\simeq 1\%$ with an aim to improve its scintillation properties.

Table 1
Summary of searches for quasi-monoenergetic solar axions coupled to nucleons through resonant excitation of nuclei.

Axion source, E_γ (keV)	Short description	lim m_a (keV)	Year [Ref.]
^{7}Li, $E_\gamma = 477.6$	HP Ge 78 cm^3, Li 61.4 g, 2667h	32.0[a]	2001 [19]
	HP Ge 160 cm^3, LiOH 3.9 kg, 3028h	16.0[b]	2005 [20]
	HP Ge 408 cm^3, LiF powder 243 g, 722h	13.9[b]	2008 [14]
	HP Ge 244 cm^3, LiF crystal 553 g, 4044h	8.6[b]	This work
^{57}Fe, $E_\gamma = 14.4$	Si(Li), Fe 33 mg (^{57}Fe 95%), 1472h	0.745[a]	1998 [21]
	Si(Li), Fe 16 mg (^{57}Fe 80%), 712h	0.360[b]	2007 [22]
	Si PIN, Fe 206 mg (^{57}Fe 96%), 334h	0.216[a]	2007 [23]
	Si(Li), Fe 290 mg (^{57}Fe 91%), 2028h	0.159[a]	2009 [24]
	Total Earth heat flux	1.6	2009 [25]
	Si(Li), Fe 1.26 g (^{57}Fe 91%), 1075h	0.145[a]	2010 [26]
^{83}Kr, $E_\gamma = 9.4$	PC[c] 243 cm^3, Kr gas 1.7 g, 564h	5.5[a]	2004 [27]

[a] At 95% C.L.
[b] At 90% C.L.
[c] Proportional counter.

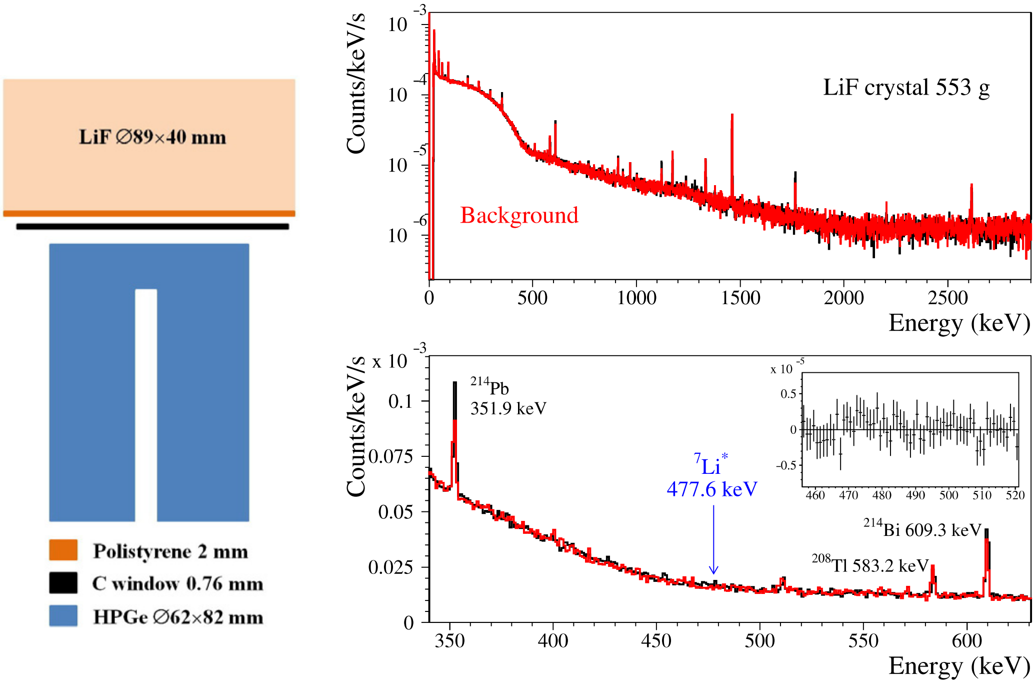

Fig. 2. (Color online.) Left: Simplified scheme of the measurements with the GeBer HP Ge detector (244 cm^3). Right: Energy spectrum measured with the LiF crystal (553 g) during 4044h (black histogram). The background spectrum (red histogram) practically coincides with the LiF data. The lower part shows the vicinity of the expected ^{7}Li peak at 477.6 keV in more detail. Difference between the rates with and without the LiF target is shown in Inset.

National Laboratories of the INFN (Italy). The detector was surrounded by a passive shield made of copper ($\simeq 10$ cm thick), low radioactivity lead ($\simeq 20$ cm), and borated polyethylene ($\simeq 10$ cm). The set-up was continuously flushed by high purity nitrogen to remove radon present in the air. The energy resolution of the spectrometer is 2.1 keV at 1332 keV γ line of ^{60}Co. The LiF target was positioned close to the HP Ge detector: distance between lower surface of the target from the detector was near 8 mm (that includes 4 mm gap between the carbon fibre window and the detector; see Fig. 2 for the simplified scheme of the measurements). The LiF sample was measured over 4043.9h (October 2008–April 2009), and the background of the detector (without the LiF target) was collected during 3046.7h (March–July 2007); the conditions of the two measurements were identical. Several other background measurements after the 2007 were carried out (during shorter periods), and the background was always at the same level. The obtained energy spectrum of the LiF sample is shown in Fig. 2 together with the background data.

The peaks found in the spectra are related with the radionuclides ^{40}K, ^{60}Co, and the daughters in the ^{232}Th and ^{238}U chains. Comparing the rates of the peaks in the spectrum accumulated with the LiF sample and in the GeBer background, one can obtain activities according to formula: $A = (S_{sample}/t_{sample} - S_{bg}/t_{bg})/(y \cdot \epsilon \cdot m)$, where S_{sample} (S_{bg}) is the area of a peak in the sample (background) spectrum; t_{sample} (t_{bg}) is the time of the sample (background) measurement; y is the yield of the corresponding γ line [13]; ϵ is the efficiency of full energy peak detection; m is the mass of the sample. Efficiencies were calculated with the GEANT4 package [28].

Only limits (at 90% C.L.) were obtained for ^{40}K (< 5.1 mBq/kg), ^{60}Co (< 0.3), ^{137}Cs (< 0.2), and for radionuclides in the ^{232}Th family: ^{228}Ac (< 1.6), ^{212}Pb (< 1.7), ^{208}Tl (< 0.6). For the ^{238}U chain, slight pollution was found as (3.3 ± 0.8) mBq/kg (^{214}Pb, ^{214}Bi). Thus, the current measurements confirmed the conclusion of the previous work [14] on the radiopurity of the LiF crystal, in contrast with the LiF powders, where the activities of U/Th daughters were as high as 0.5 Bq/kg.

3. New limit on the ^{7}Li solar axion mass

The total number of absorptions of the coupled-to-nucleons solar ^{7}Li axions through resonant mechanism in an Earth target which contains ^{7}Li nuclei was derived in [19,20] as:

$$N_{abs} = N_7 \times t \times 1.74 \times 10^{-45} \times \left(\frac{m_a}{1\ \text{eV}}\right)^4; \tag{1}$$

where N_7 is the number of ^{7}Li nuclei in a target, t is the time of measurements (in seconds), and m_a is the axion mass.

The area of the peak at 477.6 keV is equal to: $S = \varepsilon N_{abs}/(1+\alpha)$, where ε is the efficiency to detect the full energy γ with the HP Ge detector, and α is the coefficient of conversion to electrons for the given nuclear transition. Together with Eq. (1), it gives the following formula for the axion mass:

$$m_a = 1.55 \times 10^{11} \times \left(\frac{S(1+\alpha)}{\varepsilon N_7 t}\right)^{1/4}\ \text{eV}. \tag{2}$$

The full energy peak efficiency at 477.6 keV was calculated with the GEANT4 simulation package [28] as $\varepsilon = 2.27\%$. In accordance with [29], the coefficient of internal conversion for the 477.6 keV transition in ^{7}Li is extremely small: $\alpha = 7.3 \times 10^{-7}$. Taking into account the natural isotopic abundance of ^{7}Li $\delta = 92.41\%$ [30], the number of ^{7}Li nuclei in the 552.6 g LiF sample is $N_7 = 1.19 \times 10^{25}$.

As one can see from Fig. 2, the peak at the energy of 477.6 keV is absent in the LiF data, and we can give only a limit on its area with some confidence level. The value of $\lim S$ was determined in two ways. In the so-called "one σ approach" (which, notwithstanding its simplicity, gives the right scale of the experimental sensitivity), the excluded number of events that could be invisible in the spectrum is estimated simply as square root of the number of background counts in a suitably chosen energy window. Taking into account that the counting rate at energies around 478 keV is $\simeq 250$ counts/keV, and that the whole peak is not wider than $\simeq 3$ keV (FWHM value is equal $\simeq 1.6$ keV), the square root estimate gives $\lim S = 27$ events. Further, the value of $\lim S$ was determined by using the standard least squares procedure; the experimental energy distribution in the vicinity of the peak, searched for, was fitted by the sum of a linear function (representing the nearly linear background) and of two peaks: the peak being sought at 477.6 keV and the peak at 511 keV (^{208}Tl + annihilation). As a result of the fitting procedure in the energy region 460–520 keV (χ^2/n.d.f. = 0.82, where n.d.f. is number of degrees of freedom), the obtained area for the peak is (-10 ± 28) counts, giving no evidence for the effect (see Fig. 3). The maximum number of events, which can be excluded at 90% C.L., was calculated with the Feldman–Cousins procedure [31] as $\lim S = 37$. Substituting all the values in Eq. (2), one obtains the following limit on the axion mass:

$$m_a < 8.6\ \text{keV} \quad \text{at 90\% C.L.} \tag{3}$$

As one can see from Table 1, this is the most stringent limit obtained in experiments with ^{7}Li to-date.

4. Conclusions

Long-term (4044h) measurements of the radiopure LiF crystal sample with mass of 552.6 g in the underground conditions of the Gran Sasso National Laboratories (3600 m w.e.) with low background HP Ge detector (244 cm^3) lead to the new limit on the mass of the monochromatic axions coupled to nucleons, which could be emitted in the Sun in the deexcitation of the ^{7}Li nuclei and which, coming to the Earth, could excite the corresponding ^{7}Li 477.6 keV level: $m_a < 8.6$ keV at 90% C.L. This is the best limit obtained in the experiments with resonant absorption in ^{7}Li nuclei. In the used approach, the axions are coupled to nucleons both at the production and at the absorption processes, and thus the m_a limit is related only to the axion–nucleon coupling constant g_{aN}; uncertainties related with $g_{a\gamma}$ and g_{ae} disappear. The obtained limit improves the value $m_a < 13.9$ keV set in our preliminary measurements [14]. Joining the determined limit with the results of similar experiments with ^{57}Fe nuclei, one can extend a window in the excluded axion masses to the limits [477.6, 0.145] keV.

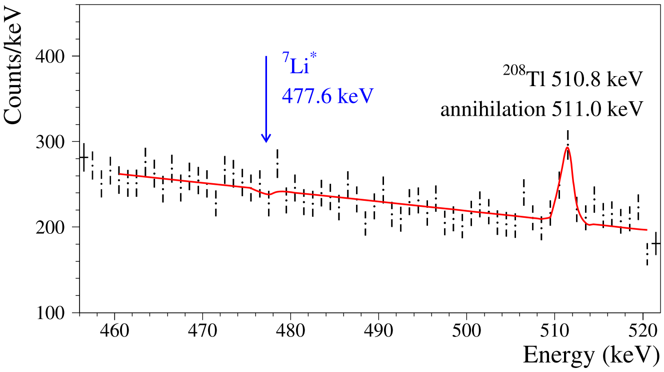

Fig. 3. (Color online.) The energy spectrum measured with the LiF sample (4044h) around the expected ^{7}Li 477.6 keV peak (points with the error bars) and its fit (continuous curve).

The present measurements confirmed the radiopurity of the LiF crystal material: only limits were obtained for ^{40}K (< 5.1 mBq/kg), ^{60}Co (< 0.3), ^{137}Cs (< 0.2), and for radionuclides in the ^{232}Th family: ^{228}Ac (< 1.6), ^{212}Pb (< 1.7), ^{208}Tl (< 0.6) at 90% C.L. For the ^{238}U chain, slight pollution was found as (3.3 ± 0.8) mBq/kg (^{214}Pb, ^{214}Bi).

Acknowledgements

The INR Kyiv group was supported in part by the Project "Kosmomikrofizyka-2" (Astroparticle Physics) of the National Academy of Sciences of Ukraine. The authors are grateful to Prof. B.V. Grinyov and Dr. V.I. Goriletsky from the Institute for Scintillation Materials (Kharkiv, Ukraine) for kindly providing the sample of LiF crystal.

References

[1] R.D. Peccei, H.R. Quinn, Phys. Rev. Lett. 38 (1977) 1440; Phys. Rev. D 16 (1977) 1791.
[2] J.E. Kim, G. Carosi, Rev. Mod. Phys. 82 (2010) 557.
[3] K. Nakamura, et al., J. Phys. G 37 (2010) 1.
[4] S. Weinberg, Phys. Rev. Lett. 40 (1978) 223.
[5] F. Wilczek, Phys. Rev. Lett. 40 (1978) 279.
[6] J.E. Kim, Phys. Rep. 150 (1987) 1.
[7] H.-Y. Cheng, Phys. Rep. 158 (1988) 1.
[8] S.J. Asztalos, et al., Annu. Rev. Nucl. Part. Sci. 56 (2006) 293.
[9] J.E. Kim, Phys. Rev. Lett. 43 (1979) 103.
[10] M.A. Shifman, A.I. Vainstein, V.I. Zakharov, Nucl. Phys. B 166 (1980) 493.
[11] M. Dine, W. Fischler, M. Srednicki, Phys. Lett. B 104 (1981) 199.
[12] A.R. Zhitnitskii, Sov. J. Nucl. Phys. 31 (1980) 260.
[13] R.B. Firestone, et al., Table of Isotopes, 8th edition, John Wiley & Sons, New York, 1996, and CD update, 1998.

[14] P. Belli, et al., Nucl. Phys. A 806 (2008) 388.
[15] L.D. Duffy, K. van Bibber, New J. Phys. 11 (2009) 105008.
[16] G.G. Raffelt, Stars as Laboratories for Fundamental Physics, University of Chicago Press, 1996;
G.G. Raffelt, J. Phys. A 40 (2007) 6607.
[17] A. Ljubicic, Rad. Phys. Chem. 74 (2005) 443.
[18] S. Moriyama, Phys. Rev. Lett. 75 (1995) 3222.
[19] M. Krcmar, et al., Phys. Rev. D 64 (2001) 115016.
[20] A.V. Derbin, et al., JETP Lett. 81 (2005) 365.
[21] M. Krcmar, et al., Phys. Lett. B 442 (1998) 38.
[22] A.V. Derbin, et al., JETP Lett. 85 (2007) 12.
[23] T. Namba, Phys. Lett. B 645 (2007) 398.
[24] A.V. Derbin, et al., Eur. Phys. J. C 62 (2009) 755.
[25] F.A. Danevich, et al., Kinemat. Phys. Cell. Bodies 25 (2009) 102.
[26] A.V. Derbin, et al., Phys. At. Nucl. 74 (2010) 596.
[27] K. Jakovcic, et al., Rad. Phys. Chem. 71 (2004) 793.
[28] S. Agostinelli, et al., Nucl. Instrum. Meth. A 506 (2003) 250;
J. Allison, et al., IEEE Trans. Nucl. Sci. 53 (2006) 270.
[29] D.R. Tilley, et al., Nucl. Phys. A 708 (2002) 3;
ENSDF at the NNDC site http://www.nndc.bnl.gov/ensdf/.
[30] M. Berglund, M.E. Wieser, Pure Appl. Chem. 83 (2011) 397.
[31] G.J. Feldman, R.D. Cousins, Phys. Rev. D 57 (1998) 3873.