

Precise measurement of the ^7Be electron capture decay half-life in silicon carbide

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Abstract

In this work we present to our knowledge the most precise measurement of the ^7Be electron capture decay half-life in a host material. A silicon carbide sample with $\sim 8.62 \times 10^9$ ^7Be atoms was measured for 83.5 d on an ultra-low background high purity Ge detector located deep underground in the Laboratori Nazionali del Gran Sasso, Italy. The result obtained for the decay half-life is $T_{1/2} = 53.284 \pm 0.016$ d, which corresponds to an uncertainty of 0.3%. Thanks to the high sensitivity achieved, this measurement is paving the way to further investigations on this process aiming to understand how environmental conditions may affect the decay half-life.

Keywords: half-life, electron capture, ^7Be



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1. Introduction

The idea of influencing the decay rate of a radioactive isotope by altering its environmental condition dates back to 1949. Segré [1] pointed out that acting on orbital electrons it is possible to modify the electron density in the nucleus which is directly connected to the half-life of electron-capture decaying atoms [2]. ${}^7\text{Be}$ is the lightest isotope decaying exclusively via electron-capture. The over-nucleus electron density modification expected with the same environmental condition is larger compared to heavier atoms. The project ASBeST (*A 7-Beryllium electron capture Study for nuclear and solid state physics*) aims to extend such investigation to the effect of a strong electric field and ionization state on the decay rate separately [3]. This article focuses on the technique developed for the study of the influence of electric field on the decay rate and the sensitivity reached. The decay of ${}^7\text{Be}$ (see figure 1) populates either the ${}^7\text{Li}$ ground state and the first excited state, followed by a gamma-ray ground state transition of energy $E_\gamma = 478$ keV that can be used for the half-life measurement. The probability for the ground state transition is $89.48 \pm 0.04\%$ and $10.52 \pm 0.04\%$ for the first excited one [4].

The currently officially adopted half-life of ${}^7\text{Be}$ is 53.22 ± 0.06 [4]. Variations from this value have been reported for several environments as reported in table 1. In [5] ${}^7\text{Be}$ has been implanted into graphite, gold, tantalum and boron nitrate leading to a variation of the order of 0.1 % in the half-life. About an order of magnitude more has been reached in [6] and [7]. In [6] ${}^7\text{Be}$ in the form of a gel ($\text{Be}(\text{OH})_2$) was exposed to high pressure. In [7] is described the last of a series of measurements (see references therein) where ${}^7\text{Be}$ is trapped into fullerene cages.

Given the subtle changes in the half-life due to environmental changes and the difficulties in making reliable predictions of the variation, all the measurements have been carried out trying to achieve the highest feasible sensitivity. In the ASBeST project, the ${}^7\text{Be}$ atoms will be implanted into the depletion region of a specifically designed silicon carbide (SiC) diode. The SiC crystal is chosen due to its high breakdown electric field ($\sim 10^8$ V m $^{-1}$) when inverse polarization is applied [8, 9]. The 4H-SiC sample is a n-type epi-layer of 5 μm thickness and $(8.8 \pm 2.2) \times 10^{15}$ cm $^{-3}$ doping and is grown on a 4 deg off-axis Si-face n-type substrate with 350 μm of thickness and 21 m $\Omega \cdot \text{cm}$ of resistivity.

The concentration of ${}^7\text{Be}$ in the semiconductor device must be kept lower than the doping one (10^{16} atoms cm $^{-3}$) not to alter the electrical properties of the device, hence the calculated electric field. Under this condition, the maximum initial ${}^7\text{Be}$ activity is of the order of 1 kBq, depending on the final size of the SiC device and on the implantation profile. In this work, we show the capability of our system to reach a precision of 0.3% in the half-life measurement, the most sensitive experiment of this kind. A SiC sample has been implanted with 8.62×10^9 ${}^7\text{Be}$ atoms (corresponding to a source activity of 1.3 kBq) and the half-life with no electric field was measured. This measurement will serve as a standard reference for the future evaluation of a possible half-life variation. In this work we use the 478 keV gamma-ray signature to evaluate the ${}^7\text{Be}$ half-life, nevertheless, also other experimental signatures can be used for the half-life estimation. For example, using the nuclear recoils and the x-ray or Auger electrons associated with the ${}^7\text{Li}$ transition, as reported in [10].

2. ${}^7\text{Be}$ implantation

The implantation was done at the 3 MV Tandem accelerator laboratory of CIRCE-DMF [11] at the University of Campania ‘Luigi Vanvitelli.’ A ${}^7\text{Be}^{2+}$ beam was accelerated to 5 MeV and implanted in the SiC crystal placed in a vacuum chamber at room temperature. The

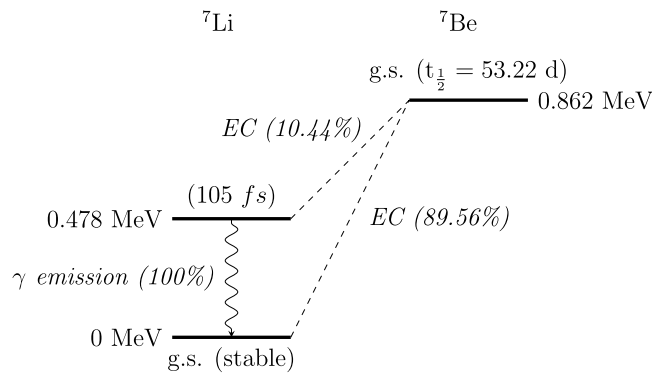


Figure 1. Decay scheme for the ${}^7\text{Be}$ nucleus from ground state.

Table 1. Summary of the ${}^7\text{Be}$ half-life measured in several materials and experimental conditions.

$T_{1/2}$ (d)	Uncertainty (d)	Host material	Activity (Bq)	References
52.68	0.05	C_{60} fullerene cage	Not specified	[7]
53.12	0.05	Beryllium metal	Not specified	[7]
53.107	0.022	Graphite	$\sim 10^5$	[5]
53.174	0.037	Boron nitride	$\sim 10^5$	[5]
53.195	0.052	Tantalum	$\sim 10^5$	[5]
53.311	0.042	Gold	$\sim 10^5$	[5]
53.414	0.003	$\text{Be}(\text{OH})_2$ gel atmospheric pressure	$\sim 10^7$	[6]
53.884	0.022	$\text{Be}(\text{OH})_2$ gel, 442 kbar pressure	$\sim 10^7$	[6]
53.22	0.06	Accepted half-life		[4]

crystal was oriented with the front face perpendicular to the beam axis to prevent the 4° channeling angle. ${}^7\text{Be}$ ions are extracted using a negative ion sputtering source MC-SNICS. The ${}^7\text{Be}$ cathode hot chemistry preparation is described in [12] and references therein. In brief, the ${}^7\text{Be}$ is extracted from the cathode in the form of ${}^7\text{Be}^{16}\text{O}^-$ molecular ion, selected through an electrostatic analyser and a dipole magnet and injected into the accelerator line. The accelerator terminal is filled with Argon, causing the molecule to break up and leading to the population of positive charge states. The $2+$ charge state of the 5 MeV beam is then selected with an analysis magnet and an electrostatic analyser. The produced beam is physiologically contaminated by ${}^7\text{Li}$, which bears the same mass and is not distinguished by the selection system. This impurity was determined using a silicon detector placed at the end of the implantation beamline, covered by a thin mylar foil. The two isobars are distinguished in the energy spectrum due to the different residual energy after passing through the foil [13]. The cathode used for the present measurement was prepared 10 months before, leading to a ${}^7\text{Li}$ contamination of over 98 %. For the purpose of this work, there were no concerns about the ${}^7\text{Li}$ contamination other than determining the actual number of ${}^7\text{Be}$ ions in the beam given the beam current, hence the duration of the implantation to achieve a ~ 1.2 kBq activity final sample. In the final device experiment, however, we will use a freshly prepared cathode to limit, as much as possible, the damage due to ${}^7\text{Li}$ implantation.

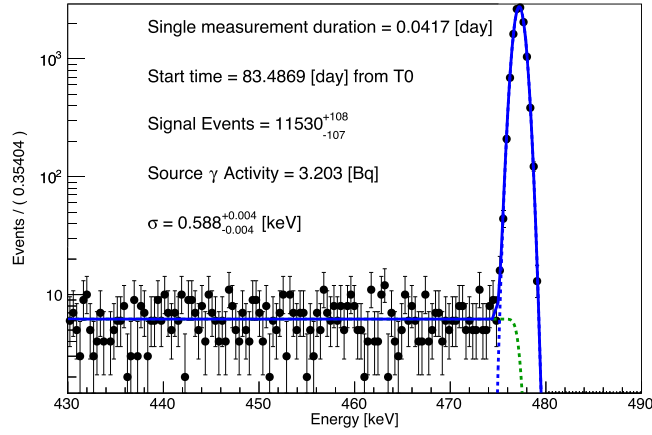


Figure 2. A typical one-hour energy spectrum acquired with the GePV detector after 83.5 d from the beginning of the data taking (T0). The blue line corresponds to the best Binned Extended Maximum Likelihood fit (BEML) obtained using an error function to reproduce the continuum Compton scattering (dashed green) plus a gaussian function with $\sigma = 0.6$ keV for the full energy peak at 478 keV (dashed blue). The gamma activity reported in the legend was evaluated dividing the number of events in the full energy peak (Signal Events) output from the best fit by the single measurement duration (3600 s). Therefore it does not represent the total activity of the ^7Be implanted source.

3. Half-life measurement: experimental setup

After the implantation the SiC sample was shipped to INFN National Laboratory of Gran Sasso (LNGS), in Italy. In order to carry out a precise measurement of the ^7Be decay half-life we used a high purity germanium (HPGe) detector in the SubTerranean Low Level Assay (STELLA) laboratory [14] located deep underground at LNGS. We take advantage of the average 1400 m rock coverage of the Gran Sasso mountain (corresponding to 3800 m.w.e. that gives a reduction factor of about one million in the cosmic ray flux) practically eliminating the contribution of the cosmic ray induced background in the HPGe detectors. Furthermore each HPGe detector is surrounded by a double layer shield: 25 cm of lead in the outer part, and about 7 cm of pure copper in the innermost part in order to attenuate the residual γ induced background coming from the natural radioactivity of the surrounding rock and concrete. This shielding is again enclosed in a one-cm-thick acrylic box, which is continuously flushed with boil-off nitrogen of the nearby liquid nitrogen storage tank to reduce as much as possible the possible presence of the radioactive noble gases ^{222}Rn and ^{220}Rn , usually omnipresent in ambient air. We placed the ^7Be implanted SiC sample inside the detector shield, directly on top of the endcap of the HPGe detector called GePV (p-type coaxial Ge detector, active volume 328 cm^3 , endcap copper, thickness 1.5 mm). We acquired data almost continuously for about 4 months; each measurement had a live time duration of 1 h. The typical energy spectrum acquired in the single measurements is shown in figure 2 with the fitted model superimposed (see caption for details). The GePV energy resolution was about 590 eV σ and that allowed to easily identify the 478 keV full energy peak from ^7Li de-excitation. Figure 2 also shows that in the time window of the single measurement the detector background was completely negligible compared to the ^7Be source activity. Furthermore, to evaluate the ^7Be half-life we studied only the trend of the number of events fully contained in the detector, i.e. the number of events under the gaussian

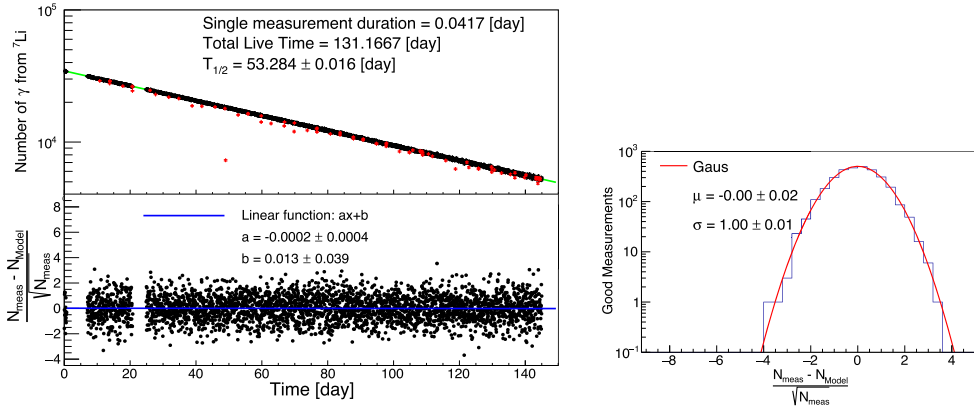


Figure 3. Left: on the top we show the number of γ -rays from ${}^7\text{Li}$ fully contained in the GePV detector as estimated from the BEML fit of the energy spectra collected (see figure 2) as a function of time. The black dots represent the 1148 accepted measurements. These data, equivalent to a live-time of 131.17 d, were used for the evaluation of the exponential decay time (green line) of the ${}^7\text{Be}$ decay. The measurements where we observed detector instabilities were rejected from the half-life evaluation and are marked in red. Each data point has as y and x error bars, the asymmetric ones obtained from the UEML fit (see in figure 2 Signal Events) and the measurement time divided $\sqrt{12}$, respectively. On the bottom we show the residuals $((N_{\text{meas}} - N_{\text{model}})/\sqrt{N_{\text{meas}}})$ between the accepted measurements and the best fit exponential shape. The blue line represents one degree polynomial fit of the residuals as a function of time: both the function parameters (a and b) are compatible with zero. Finally, on the right-hand plot, the residuals' distribution was fitted with a gaussian function which returns the expected values of μ and σ (0 and 1 respectively).

depicted in figure 2 as a function of time. In such a way we further reduced the background events coming from the residual environmental radioactivity since the energy range of interest was only 3 keV wide and, thus, the expected rate of background events in this region in one hour of measurement was lower than 0.05 h^{-1} .

4. Half-life measurement: data analysis

In figure 3 on the left we report the trend of the number of 478 keV γ -rays from ${}^7\text{Li}$ fully contained in the GePV detector as a function of time.

We had two discontinuities in the data taking caused by DAQ software instabilities, the first one after few hours of data taking and the second one about 20 d after, as shown in figure 3. We collected in total 3230 single measurements, 82 of them were rejected because of detector instabilities caused by the periodic filling of the detector dewar with liquid nitrogen. These measurements are reported in figure 3 (left) (the points flagged in red), but are not used for the ${}^7\text{Be}$ half-life extrapolation. The points in black with both x and y associated uncertainties were fitted with an exponential function with only two parameters using ROOT [15], the exponent (τ) and a constant term. The corresponding half-life value ($T_{1/2} = \ln(2)/\tau$) is $53.284 \pm 0.016 \text{ d}$. With a precision of 0.3% this estimation results to be the most accurate measurement of the ${}^7\text{Be}$ half-life reported in literature [7]. As crosscheck we also studied the residual distribution (see figure 3 right) and its trend as a function of time (see figure 3 left bottom). It is constant over time, which proves that a single exponential function well

Table 2. Summary of the study of the systematic uncertainty contribution to the ${}^7\text{Be}$ half-life.

Systematic source	$T_{1/2}$ (d)	Statistical (d)
No systematic	53.284	± 0.016
1 s detector dead time	53.284	± 0.016
Flat bkg contribution	53.284	± 0.016
UEML fit procedure	53.280	± 0.016

reproduces the collected data. Finally the distribution is fully compatible with a gaussian with average zero and unitary sigma as expected for a good residuals distribution.

5. Systematic uncertainty

On the previous section we illustrated the data analysis of the collected measurements. The uncertainty showed accounts only for the statistically one. In this section we discuss the possible contribution of the systematic uncertainties on the half-life measurements as discussed in [16] and in the reference therein: background subtraction, dead time compensation, geometrical changes, non reproducibility in the detection efficiency due to various conditions, imperfect fit of peak and continuum with changing spectral shapes. Concerning the background subtraction we take advantage of the ultra low background level of the detector located in the STELLA facility: as underlined at the end of section 3 the background rate measured in the energy region used to evaluate the number of gamma collected coming from the ${}^7\text{Be}$ decay is lower than $0.05 \text{ count h}^{-1}$. On the other hand the signal is of the order of $10^4 \text{ count h}^{-1}$. Then the number of counts coming from background is 2000 time lower than the statistical uncertainty ($\sqrt{10^4} = 10^2$) due to the number of collected signal events. For that reason we did not perform any background subtraction in the collected energy spectra. Nevertheless we investigate a possible contribution coming from such background fitting the data points collected with an exponential function plus a constant background contribution limited between 0 and 0.05 counts h^{-1} : ($N_0 e^{(-t/\tau)} + BKG$). No differences were found. The ultra low environmental background and the low activity of the ${}^7\text{Be}$ source allowed us also to minimized the possible systematic uncertainty coming from dead time compensation. Indeed, in a HPGe detector the dead time arise only from high counting rate because of pile-up events. Following the calculation in [17] we estimate a dead time of 0.02%–0.03%, i.e. less than a second in one hour. Then, we repeated the half-life evaluation using 3599 s instead of 3600 s as time duration for the single measurement and no variation in the fit results was observed.

Concerning the detector efficiency to γ radiation it depends on the γ energy, the source position and dimension and on the detector dimensions and density. All these parameters remain unchanged for all the measurements period. Then we do not expect any variation on the detector efficiency that can affect the collected data. We finally investigated the possible systematic uncertainty coming from the BEML fitting procedure of the ${}^7\text{Be}$ full energy peak plus the continuum due to Compton scattering. To study this effect we repeated the data analysis using roughly the number of event collected in the energy range of $476 \div 480 \text{ keV}$ without performing any fit, and associated to each point the symmetric y uncertainty of $\pm \sqrt{N}$ (the x uncertainty remained the same): the ${}^7\text{Be}$ half-life obtained using this approach was $53.280 \pm 0.016 \text{ d}$. In conclusion all the possible source of systematic uncertainties have been taken into account (as summarized in table 2) and for all of them we obtained a null value or a deviation from the result shown in section 4 smaller than the statistical uncertainty of 0.016 d.

6. Conclusion

In this paper we demonstrate that using a ~ 1.3 kBq ^7Be source implanted into a SiC sample and placed on to an ultra-low background HPGe detector we are able to achieve a very good accuracy in the measurement of the ^7Be decay half-life of the order of 0.3%. The possible contribution coming from systematic uncertainty was taken into account and it resulted in -0.004 d, i.e. subdominant with the respect to the statistically one. This measurement is the most precise one for the ^7Be electron capture decay half-life in literature. Using the same procedure we will be able to observe deviations $\geq 1\%$ depending on the environmental condition where the decay occurred.

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Data availability statement

The data cannot be made publicly available upon publication because they are not available in a format that is sufficiently accessible or reusable by other researchers. The data that support the findings of this study are available upon reasonable request from the authors.

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