New measurement of ⁷Be half-life in different metallic environments

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Abstract. ⁷Be decays via electron capture and therefore its half-life is expected to depend on the electron density at the nucleus. We measured the ⁷Be half-life in palladium, tungsten, zirconium and tantalum metals, in order to investigate the influence of the quasi-free electrons in metals on the probability of electron capture. The ⁷Be samples were obtained implanting a 4.85 MeV pure radioactive ⁷Be ion beam. The ⁷Be half-life was determined measuring the 478 keV gamma decay following the electron capture by means of HPGe detectors. In order to reduce systematic errors, we planned to perform independent measurements in three different laboratories: Naples (Italy), Bochum (Germany) and Debrecen (Hungary). On the basis of the first results, we do not find a ⁷Be half-life change within the experimental errors of 0.4%.

PACS. 21.10.Tg Lifetimes – 23.40.-s β decay; double β decay; electron and muon capture – 27.20.+n $6 \le A \le 19$

1 Introduction

In the stellar environment, ⁷Be is produced by ${}^{3}\text{He}(\alpha,\gamma){}^{7}\text{Be}$ and destroyed by ${}^{7}\text{Be}(p,\gamma){}^{8}\text{B}$, which is responsible for the high-energy solar neutrino flux, and, in competition, by ⁷Be decay. ⁷Be decays to ⁷Li via electron capture, populating either the ⁷Li ground state or, in 10% of the cases, the first excited state, which decays to the ground state emitting a 478 keV gamma ray.

The half-life of the nuclei such as ⁷Be which decays by electron capture depends on the electron density at the nucleus. Therefore, the decay probability of these nuclei in stellar plasma environment differs from their terrestrial values. Starting from 1949 [1], many experiments were done in order to measure variations of the ⁷Be half-life depending on the host chemical environment. Although the half-life changes are in most cases lower than 0.2% [2], some authors found larger variations [3,4,5].

Recently, it was discovered that the electron screening in d(d, p)t reaction for deuterated metals is much higher than the screening measured in d(d, p)t using deuterium gas target [6]. This feature has been explained by the Debye plasma model applied to the quasi-free electrons in metals: these electrons form an electron cloud around the nucleus with a Debye radius which is about a factor 10 smaller than the atomic radius [7]. According to this picture, ⁷Be nuclei implanted in metallic host materials may probe a higher electron density, that would provide a test case for the calculations of the ⁷Be decay in the solar plasma.

We implanted ⁷Be in different metallic targets and we measured the half-life of an electron capture in order to highlight a detectable half-life variation depending on the different quasi-free electronic density. The choice of the host materials was based on the measurements of the electron screening potential for d(d, p)t in deuterated metals [6]: palladium, tungsten and tantalum metals show a large screening potential, whereas for zirconium the screening effect is smaller.

In previous experiments, with the exception of [8], secondary ⁷Be beams have been used and therefore both the projectile and the ⁷Be recoil have been implanted at roughly the same depth. As a consequence, the electron distribution around ⁷Be nuclei may be altered due to a) the chemical contamination of the beam; b) the lattice damage induced by the high implantation currents. In this experiment, we use a different approach, *i.e.* a pure ⁷Be beam is implanted in host materials in order to virtually reduce to zero the implantation damage and avoid changes of the sample stoichiometry. Table 1 shows the experimen-

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Author	Primary beam	Host material, $^7\mathrm{Be}$ range	⁷ Be half-life (d) or ratio (%)
F. Lagoutine [8]	⁷ Be (Energy not reported)	metallic Al, –	53.17 ± 0.07
E.B. Norman [9]	$40 \mathrm{MeV}$ ³ He	grafite, -	53.107 ± 0.022
	$10 \mathrm{MeV}$ H	boron nitride, –	53.174 ± 0.037
	$45 \mathrm{MeV}$ ⁷ Li	Ta, -	53.195 ± 0.052
		Au, -	53.311 ± 0.042
T. Ohtsuki [3]	$16 \mathrm{MeV}$ H	C_{60} cages, –	52.68 ± 0.05
	γ irradiation	metallic ⁹ Be, –	53.12 ± 0.05
A. Ray [4]	$7 \mathrm{MeV}$ H	Au, $1.8\mu\mathrm{m}$	$(T_{1/2}(Au) - T_{1/2}(Al_2O_3))/T_{1/2}(Al_2O_3)$
		$Al_2O_3, 2\mu m$	$= (0.72 \pm 0.07)\%$
Zhou $[5]$	$^{7}\text{Li}/^{7}\text{Be}$ mixed beam	Pd, $23.6\mu\mathrm{m}$	$(T_{1/2}(Au) - T_{1/2}(Pd))/T_{1/2}(Pd)$
		Au, $20.9\mu\mathrm{m}$	$= (0.8 \pm 0.2)\%$
Liu [<mark>10</mark>]	$3.2\mathrm{MeV}$ H	natural beryllium, $2\mu m$	$(T_{1/2}(Au) - T_{1/2}(Be))/T_{1/2}(Be)$
	-	natural gold, $1\mu m$	$\leq 0.12\%$
Present work, [11]	⁷ Be	$\mathrm{Pd},2.9\mathrm{\mu m}$	see table 3
		$ m W,2.7\mu m$	
		$Zr, 4.3 \mu m$	
		Ta, $3.1 \mu\text{m}$	

Table 1. Implantation procedure of earlier ⁷Be half-life measurements.

tal procedure of the $^7\mathrm{Be}$ implantation of the earlier and present $^7\mathrm{Be}$ half-life measurements.

Moreover, the ⁷Be half-life has been independently measured in 3 different laboratories in order to reduce the effects of possible systematic errors.

2 Experimental procedure

The production of a pure ⁷Be beam at the 3MV Tandem accelerator in Naples is described elsewhere [12]. Shortly, ⁷Be is produced via ${}^{7}Li(p, n){}^{7}Be$ using a 11.7 MeV proton beam with a current intensity of $20 \,\mu\text{A}$, delivered by the ATOMKI Cyclotron of Debrecen (Hungary). ⁷Be is then extracted from the metallic Li target by means of radiochemical methods [12] at the isotopic laboratory of the Ruhr-Universität Bochum (Germany). The activity is collected into a cathode of the sputtering ion source which is used to produce a ⁷BeO⁻ beam. This molecular ion beam is injected in the accelerator and the emerging $^{7}\text{Be}^{2+}$ beam is selected as a ${}^{7}\text{Be}^{4+}$ beam, after passing a thin C foil. This procedure suppresses the Li contamination in the beam and allows to obtain a high-purity ⁷Be beam. The beam intensity was about 7 ppA. A pair of scanning magnets provided a uniform implantation. The scheme of the experimental setup is shown in fig. 1.

The implanted metallic materials are Pd, W, Zr, Ta with activities ranging from 3.6 to 36 kBq. The kinetic beam energy $E_{lab} = 4.85$ MeV provided about 3–4 μ m ion stopping range (table 1), which is deeper than the layers where one expects surface contaminations and oxidation of the metal. The ⁷Be half-life of the implanted samples was independently measured in the laboratories of Bochum, Debrecen and Naples. The 478 keV gamma emission was detected with HPGe detectors. A ⁶⁰Co source was included in all measuring setups (except for the W Naples

Table 2. Experimental details of the Debrecen and Naples ⁷Be half-life measurements.

	Debrecen	Naples
HPGe Detector efficiency		
$(3'' \times 3'')$ NaI scale %)	20	28
Pb shield thickness (cm)	5	5
Sample to HPGe distance (cm)	3	0
60 Co to HPGe distance (cm)	3	10
Analyzed samples	Pd-W	Pd-W
	Zr-Ta	
γ spectra time		
integration (h)	12	1-6
Measuring time (days)	37 - 40	50 - 100
	74 - 37	

measurement). In this work we show the first experimental results of Debrecen and Naples groups. The details of these two setups are summarized in table 2.

The γ spectra were automatically stored at every 1, 6 or 12 hours, depending on the activity of the sample. As an example, fig. 2 shows a typical spectrum containing the ⁷Be decay peak, 511 keV annihilation peak and the two γ lines of ⁶⁰Co reference source.

In principle, possible sources of systematic errors are: a) efficiency variation during these long measurements; b) pileup effect which may increase the half-life value; c) acquisition dead time; d) absolute time shifts. The a), b), c) errors can be avoided if the half-life value is evaluated by normalising the ⁷Be counts to another independent source. To this purpose, we used two ⁶⁰Co peaks and the 1460 keV natural background peak of ⁴⁰K. As regards d), the error was kept below 0.01%. For example, the internal clock of the acquisition PC of Naples setup was compared at the beginning of each run to the Greenwich



Fig. 1. Scheme of the ⁷Be implantation setup at the TTT3 Tandem accelerator of the Federico II University of Naples.



Fig. 2. Gamma spectrum of Pd sample measured in Naples $(\Delta t = 1 \text{ h})$.

reference time (National Institute of Standards and Technology web site: www.tf.nist.gov/service/its.htm).

Finally, it should be noted that the choice of the experimental setup, the data acquisition and the analysis procedure were completely left to the different groups which independently draw out their results.

3 Results

The samples measured in Naples were Pd, W, Zr. The data analysis was completed for the Pd and the W targets, whereas the analysis of the Zr sample is still in progress. The complete data analysis of Debrecen workgroup is available for all the implanted targets W, Pd, Zr and Ta. All the results are shown in table 3. As an example, the details of data analysis at Naples are presented below.

Every spectrum has been analysed fitting a linear background below the ⁷Be and the reference peaks. We obtain a set of values $\{A_n^{exp} \pm \sigma_n\}$ which are the time integrals of the peaks after a linear background subtraction. This analysis has been done for both ⁷Be and the reference peaks. A least-square function Q^2 ,

$$Q^2 = \sum_{n} \left[\frac{A_n^{exp} - A_n^{th}}{\sigma_n} \right]^2, \tag{1}$$

was minimized using the experimental values A_n^{exp} and the fit function A_n^{th} :

$$A_n^{th} = \int_{start}^{stop} A_0 \epsilon e^{-t/\tau} dt - \int_{(\Delta t)_{dead}} A_0 \epsilon e^{-t/\tau} dt$$
$$= \int_{start}^{start + \Delta t_{live}} A_0 \epsilon e^{-t/\tau} dt$$
$$\epsilon = const}{}_{a_0 \epsilon \tau e^{-t_{start}/\tau}} (1 - e^{-\Delta t_{live}/\tau}), \qquad (2)$$

where A_0 is the initial activity, ϵ is the detection efficiency and Δt_{live} and Δt_{dead} are the acquisition live and dead time, respectively. The free parameters are $(A_0\epsilon)$ and τ . The uncertainty on τ is determined by means of a χ^2 analysis.

Similar expressions were used for the ratio with the reference sources.

For each implanted sample, we evaluated the ⁷Be halflife by fitting both the ⁷Be counts and the ratio between

	Workgroup		⁷ Be half-life [d]		
Normalization method		Pd	W	Zr	Та
Without normalization	Naples	53.14 ± 0.05	53.65 ± 0.06		
	Debrecen	53.28 ± 0.14	52.95 ± 0.15	53.01 ± 0.05	53.21 ± 0.34
	Average	53.16 ± 0.05	53.55 ± 0.06		
Norm. to $1173{\rm keV}^{-60}{\rm Co}$ line	Naples	53.13 ± 0.14	-		
	Debrecen	53.18 ± 0.22	53.32 ± 0.21	53.04 ± 0.15	52.67 ± 0.36
	Average	53.14 ± 0.12			
Norm. to $1332 \mathrm{keV}^{-60}$ Co line	Naples	53.31 ± 0.14	-		
	Debrecen	53.30 ± 0.23	53.07 ± 0.21	53.02 ± 0.15	52.70 ± 0.36
	Average	53.31 ± 0.12			
Norm. to $1460 \mathrm{keV}^{40} \mathrm{K}$ line	Naples	_	54.72 ± 0.94		
	Debrecen	53.41 ± 0.48	53.39 ± 0.43	53.12 ± 0.20	53.46 ± 0.51
	Average		53.62 ± 0.39		

Table 3. First experimental results of Debrecen and Naples groups. The 7 Be half-life values evaluated with and without normalization are reported.

⁷Be and the reference source peaks. The half-life values obtained without normalization are in agreement with the normalized ones. The values and the errors evaluated using the normalized data take into account some systematic errors like the pileup effect, the efficiency variation and the acquisition dead time. A direct comparison between Naples and Debrecen data is possible for Pd and W metals. Unfortunately, for the W metal, the precision of the Naples measurement is not sufficient, since no additional reference source was available in the experimental setup.

The results have to be compared to the ⁷Be half-life adopted value [2] of 53.22 ± 0.06 days. Although the data analysis has to be finalized for all the implanted metals, on the basis of the first Debrecen and Naples data above, we do not find ⁷Be half-life change within the experimental errors (0.3-0.4%).

This result is compatible at a 2σ level with the predictions of the Debye model applied to the free electrons in metals, provided the following additional assumptions are made: a) the contribution of the free electrons capture is negligible; b) the Debye screening does not affect the electron steady wave functions of ⁷Be atoms. In this case, in fact, the ⁷Be screening potential U_D due to the free electrons is $U_D = -1 \times 4 \times U_{d+d}$, where 4 is Beryllium atomic number and U_{d+d} is the electron screening value measured in d(d, p)t reaction in metallic environments. Between the metals considered, the highest U_{d+d} value is 800 eV for Palladium [6], which gives $U_D = -3.2 \text{ keV}$. Since the electron capture process scales with the energy squared, the reduction factor of the decay probability is $[(862-3.2)/862]^2 = 0.992$, *i.e.* 0.8%, which correspond to a longer half-life $(^1)$.

Finally, our results seem not to confirm the findings of [5] where a correlation between the screening and the ⁷Be half-life was observed having opposite sign respect to the predictions of the Debye model. However, a realistic model of the electron capture decay of ⁷Be in metallic environment should be developed to compare these experimental results with the data of the electron screening in metals.

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 $^{^1}$ The energy release of the $^7\mathrm{Be}$ electron capture decay is 862 keV.