

Change of the ${}^7\text{Be}$ electron capture half-life in metallic environments

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Abstract. For the electron capture of ${}^7\text{Be}$ in the metallic environments Pd and In the ${}^7\text{Be}$ half-life was observed to increase by 0.9 ± 0.2 and $0.7 \pm 0.2\%$, respectively, while in the insulator Li_2O it was unchanged within experimental error (all samples cooled to $T = 12\text{ K}$). The observations are consistent with the predictions of the Debye plasma model applied to the quasi-free electrons in the metals.

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The cross-section of a nuclear fusion reaction is enhanced at sub-Coulomb energies by the electron clouds surrounding the interacting nuclei, with an enhancement factor [1, 2]

$$f_{\text{lab}}(E) = E(E + U_e)^{-1} \exp(\pi\eta U_e/E), \quad (1)$$

where E is the center-of-mass energy, η the Sommerfeld parameter, and U_e the electron screening energy. Recently, the electron screening in d(d,p)t has been studied for deuterated metals and insulators, *i.e.* 58 samples in total [3–5]. As compared to measurements performed with a gaseous D_2 target ($U_e = 25 \pm 5\text{ eV}$ [6]; theory = 27 eV), a large screening was observed in all metals of order $U_e = 300\text{ eV}$, while a small (gaseous) screening was found for the insulators. An explanation of the large screening in metals was suggested by the Debye plasma model applied to the quasi-free metallic electrons. The electron Debye radius around the deuterons in the lattice is given by

$$R_D = (\varepsilon_0 kT / e^2 n_{\text{eff}} \rho_a)^{1/2} = 69(T / n_{\text{eff}} \rho_a)^{1/2} \quad [\text{m}], \quad (2)$$

with the temperature T of the quasi-free electrons in units of K, n_{eff} the number of these electrons per metallic atom, and the atomic density ρ_a in units of atoms/m^3 . With the Coulomb energy of the Debye electron cloud and a deuteron projectile at R_D set equal to $U_e \equiv U_D$, one obtains

$$U_D = 2.09 \times 10^{-11} (n_{\text{eff}} \rho_a / T)^{1/2} \quad [\text{eV}]. \quad (3)$$

A comparison of the calculated and observed U_e values led to n_{eff} values, which were compared with those from the Hall coefficient: they agreed within experimental uncertainties. Another test was the predicted temperature dependence, $U_D \propto T^{-1/2}$, which was verified for $T = 260$ to 670 K . Furthermore, the Debye energy U_D should scale with the nuclear charge Z_t of the target atoms and the charge Z_i of the incident ions, $U_D \propto Z_t Z_i$; these predictions were also verified [7–9].

There is another prediction of the Debye model concerning radioactive decay of nuclides in a metallic environment. In general, for the α -decay and β^+ -decay one expects a shorter half-life due to the acceleration mechanism of the Debye electrons for these positively charged particles, while for the β^- -decay and electron capture pro-

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cess one predicts correspondingly a longer half-life. For example, if the electron capture by ${}^7\text{Be}$ ($Q = 862\text{ keV}$; $T_{1/2} = 53.29\text{ d}$) occurs in a metal cooled to $T = 12\text{ K}$, one arrives at $U_D = Z_e Z_t U_e (d + d)(293/12)^{1/2} \approx 1 \times 4 \times 300\text{ eV} \times 4.94 = 5.9\text{ keV}$, where we used a typical value of 300 eV for the $d + d$ fusion reaction at $T = 293\text{ K}$ and assumed the $U_D \propto T^{-1/2}$ dependence to be valid for $T < 260\text{ K}$. A possible temperature dependence of $n_{\text{eff}}(T)$ is not known from the Hall coefficient and has thus been neglected. Since the lifetime scales with the energy squared, one obtains $f_{\text{lab}} \approx ((Q - U_D)/Q)^2 = 0.986$, *i.e.* a longer half-life by 1.4% . At room temperature the half-life increases by 0.3% , consistent with recent measurements ([10] and references therein). Thus, one expects a change in lifetime of 1.1% between $T = 293$ and 12 K . If the Debye electrons also contribute significantly to the capture process, one expects here a shorter half-life; thus both effects may cancel each other. The present work reports on experimental investigations of the lifetime of ${}^7\text{Be}$ in metallic and non-metallic environments cooled to $T = 12\text{ K}$.

The setup consisted of a cryopump (Leybold RGD210, minimum fixed temperature $T = 12\text{ K}$), where on the head of its Cu rod (length $l = 30\text{ cm}$, thermal expansion coefficient for Cu, $\alpha = 1.65 \times 10^{-5}\text{ K}^{-1}$) the sample was installed together with a Si diode (Lake Shore Cryotronics DT-670) for the temperature measurement. The chamber with the cryopump was evacuated by a turbopump (pressure = $2 \times 10^{-9}\text{ mbar}$ with the cryopump on). Since the electron capture of ${}^7\text{Be}$ proceeds to 10% via the 478 keV first excited state of ${}^7\text{Li}$, we used a Ge detector placed at 0° to the cryopump axis (100% relative efficiency, resolution = 1.5 keV at $E_\gamma = 478\text{ keV}$, front-face distance to the sample $d = 8\text{ cm}$) to observe the 478 keV γ -rays. The setup was surrounded by a 5 to 10 cm thick Pb shield to suppress the room background. A 50 Hz pulser was used to check for dead time effects ($< 0.1\%$). The ${}^7\text{Be}$ sample required the following fabrication steps: i) the ${}^7\text{Be}$ nuclides were produced via the reaction ${}^7\text{Li}(p, n){}^7\text{Be}$ at the cyclotron in Debrecen; ii) the target was enriched in ${}^7\text{Be}$ via hot chemistry at the Isotopenlabor in Bochum [11]; iii) the produced ${}^7\text{Be}/{}^7\text{Li}$ pill was installed in the sputter ion source of the 3 MV Tandetron in Caserta producing a pure ${}^7\text{Be}^{4+}$ beam of 6.5 MeV with a beam intensity of about 2×10^5 ${}^7\text{Be}/\text{s}$; iv) the ${}^7\text{Be}$ beam was implanted in the metals Pd and In. From the ${}^7\text{Be}/{}^7\text{Li}$ material we produced a ${}^7\text{BeO}$ sample in a Li_2O matrix (insulator) and mixed the matrix also with ${}^{137}\text{Cs}$ nuclides (backing = Si wafer). A ${}^7\text{Be}$ γ -activity of 200 to 500 Bq was achieved for the samples. The experimental procedure consisted of 2 steps: i) starting at room temperature $T(t = 0) = 293\text{ K}$ we measured the relative 478 keV γ -ray activity $A(t = 0)$ over a running period of 24 to 48 h depending on the activity of the sample; ii) the cryopump was then turned on, where the temperature limit $T = 12\text{ K}$ was reached in 3 h ; we measured then the relative 478 keV γ -ray activity $A(t)$, where again a long run was taken (24 to 48 h). The cryopump was then turned off, where room temperature was reached in 12 h . The 2 steps have been repeated if statistical accuracy was not sufficient.

The same procedure was followed using radioactive sources, in order to investigate the dependence of the detection efficiency on the temperature. First, we used a ${}^{22}\text{Na}$ source ($E_\gamma = 511\text{ keV}$, $T_{1/2} = 2.602\text{ y}$) and observed a change of the activity (corrected for the normal $\exp(-t/\tau)$ radioactive decay) by $-1.2 \pm 0.1\%$ between $T = 293$ and 12 K . From the mixed ${}^7\text{BeO} + {}^{137}\text{Cs}$ ($E_\gamma = 662\text{ keV}$, $T_{1/2} = 30.17\text{ y}$) sample, the 662 keV γ -ray flux led to the same change of the activity: $-1.2 \pm 0.3\%$. For the given geometry and the thermal contraction $\Delta l = 0.70\text{ mm}$ (leading to an increase in the distance d and thus to a decrease of the solid angle), GEANT simulations resulted in a value of $-1.2 \pm 0.1\%$ for $E_\gamma = 478\text{ keV}$. We adopted the value $-1.2 \pm 0.1\%$ for the correction of the detection efficiency due to the thermal contraction of the Cu rod. With the ${}^7\text{Be}$ implanted samples of Pd and In and the ${}^7\text{BeO}/\text{Li}_2\text{O}$ mixed matrix placed —each at a time— at the head of the cryopump we found a change of the 478 keV γ -ray activity by -1.7 ± 0.2 , -1.9 ± 0.2 , and $-1.0 \pm 0.2\%$, respectively, where the normal $\exp(-t/\tau)$ decay has been taken again into account. Correcting the results for the thermal contraction, we arrive for the metals Pd and In at a longer ${}^7\text{Be}$ half-life by -0.5 ± 0.2 and $-0.7 \pm 0.2\%$, respectively, while for the insulator Li_2O there is no change of the half-life within experimental error ($+0.2 \pm 0.3\%$).

Measurements of the ${}^7\text{Be}$ -Pd sample have been repeated with the Ge detector placed at 90° to the sample holder, where the normal of the sample ($d = 12\text{ cm}$) was oriented perpendicular to the axis of the cryopump. In this arrangement the effect of thermal contraction of the Cu rod is expected to be sizably reduced. The result for the ${}^7\text{Be}$ activity is a change by -1.1 ± 0.2 , while the ${}^{22}\text{Na}$ source led to $+0.1 \pm 0.1\%$ confirming the expectation. After correction for the source value, we obtain $-1.2 \pm 0.2\%$ for Pd.

As final results we adopted the weighted average of $-0.9 \pm 0.2\%$ (Pd) and $-0.7 \pm 0.2\%$ (In). The values for the longer ${}^7\text{Be}$ lifetimes are in principle consistent with expectation from the Debye model (see above) but they are somewhat smaller than expected for a capture process by the atomic electrons alone and may indicate therefore a significant additional contribution by the Debye electrons themselves (*i.e.*, continuum electron capture [2]); estimates indicate a negligible contribution. Thus, the smaller value may have an experimental origin requiring the use of ultra-clean metals with oxygen and hydrogen contaminations of less than 1 ppm (*e.g.*, Cu or Au). Finally, it appears desirable to have a cryo-setup with an adjustable temperature; such a setup is in preparation.

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