Towards a measurement of α-decay lifetime variation at high pressure: The case of ^{228}Th

Fabio Belloni[1], Noaz Nissim[2], Vincenzo Romanello[3], Shalom Eliezer[4,5]

[1] *European Commission, Directorate-General for Research and Innovation, Directorate Energy, Brussels, Belgium; email: fabio.belloni@ec.europa.eu*
[2] *Department of Earth and Planetary Science, University of California Berkeley, California, USA*
[3] *Research Centre Rez, Husinec-Rez 130, Czech Republic*
[4] *Soreq Nuclear Research Centre, Yavne, Israel*
[5] *Institute of Nuclear Fusion, Polytechnic University of Madrid, Madrid, Spain*

Abstract

We suggest that a change in the lifetime of the α decay of ^{228}Th ($T_{1/2} = 1.9127(5)\ \mathrm{yr}$) may be detected at static pressures as high as 1 Mbar, a regime nowadays achievable in the laboratory by means of diamond anvil cells. The effect is due to the perturbation of the nuclear Coulomb potential and decay *Q*-value by the electron environment, and is enhanced, in this case, by the high compressibility of Th. The Thomas-Fermi model, within the frame of the Feynman-Metropolis-Teller treatment of the equation of state, has been used for calculations. It is found that at a compression factor of 2 on the volume of the Wigner-Seitz cell, the fractional variation in the lifetime of ^{228}Th is about -2×10^{-4} (conservative estimate). Detailed experimental procedures to measure this effect by compressing ^{228}Th metal in a diamond anvil cell are presented, where diagnostics is based on activity-*vs*-time recording on suitable lines of the decay γ-spectrum, *e.g.* the 2.614-MeV peak stemming from the disintegration of the decay product ^{208}Tl, at secular equilibrium.

Introduction

It is customary to treat the lifetime of radioactive elements as a constant of nature, although already in the mid-20th century Segré [1] and Daudel [2] conjectured that it might be dependent on environmental conditions such as the chemical composition of the host matrix, temperature, and pressure. So far, the clearest evidence obtained in condensed matter mainly concerns the decay of ^{7}Be (electron capture) [3,4] and ^{99m}Tc (internal conversion) [5] at high pressure (hundreds kbar), with variations of the order of 10^{-2} and 10^{-4}, respectively (indeed, atomic electrons contribute directly to these decay modes). On the basis of considerations on time-reversal symmetry of *screened* charged-particle nuclear reactions, α and β decays have recently been proposed to also be sensitive to the effect of the electron environment [6,7], which has renewed interest in the possibility of manipulating their lifetime.

For α decay, in particular, it has been noted that the effect could have implications for the stellar nucleosynthesis of heavy elements and nuclear cosmochronology [6,8,9] as well as applications to the determination of the screening enhancement factor in nuclear reactions of astrophysical interest [9,10]. Possible applications to nuclear waste management have also been the subject of speculation, in connection to the supposed increase, by several orders of magnitude, of the decay rate in a metal lattice at low temperature [6,11]. A much more modest experimental evidence (below 10%) subsequently obtained on ^{210}Po in a Cu matrix at $T = 12$ K [12] has however been disproven [13].

More recently, the effect of compression on α-decay has been studied [14,15]. It has been found that a measurable lifetime variation can be achieved at static compressions as high as those induced by means of diamond anvil cells (DACs). Lately, we have assessed the feasibility of a measurement on the highly compressible ^{241}Am metal, at DAC pressures [16]. We have shown that the small extent of the effect (fractional half-life reduction of 2 x 10^{-4} at 0.5 Mbar) together with the relatively long half-life of ^{241}Am make methods based on γ activity inapplicable, the only viable option being a mass-spectrometry measurement of the variation of the ^{237}Np/^{241}Am ratio upon compression, after a decay time of the order of 1 yr.

Following the screening of a number of candidate nuclides, in this paper we propose a better measurement of the effect, based on activity-*vs*-time monitoring via γ spectrometry, on the α decay of DAC-compressed ^{228}Th. The decay chain of ^{228}Th ($T_{1/2} = 1.9127(5)$ yr) is composed of several short-lived products rapidly reaching secular equilibrium [17]. The resulting rich γ-spectrum (Fig. 1), combined to the good compressibility of Th [18] and the relatively short half-life, indeed make ^{228}Th metal possibly the best option for this kind of measurement.

Theory

In the customary one-body model of the *bare* decay, the daughter nucleus and the α particle interact through a potential, V_b, dependent only on their relative distance *r*;

$$V_b(r) = V_N(r) + V_C(r) + V_\ell(r) \quad (1)$$

where V_N is the attractive intra-nuclear potential, V_C is the Coulomb potential, and V_ℓ is the centrifugal potential associated to the relative angular momentum ℓ. We have here adopted the model of Buck *et al.* [19] for the calculation of V_b and of the decay width, Γ_b, as explained in ref. [15]. In this formalism,

$$\Gamma_b = (\hbar^2/4\mu) P_b F_b exp(-2G_b) \quad (2)$$

where μ is the reduced mass of the system, P_b is the α-particle preformation probability, the Gamow factor G_b is given by $G_b = \int_{r_1}^{r_2} k_b(r)dr$, and the normalization factor F_b by $F_b^{-1} = (1/2)\int_{r_0}^{r_1} k_b^{-1}(r)dr$. The wavenumber k_b is given by

$$k_b = \sqrt{(2\mu/\hbar^2)|V_b(r) - Q_b|} \quad (3)$$

where Q_b is the total kinetic energy released in the decay; the classical turning points r_0, r_1, r_2 are the solutions of the equation $k_b(r) = 0$. The decay constant, λ_b, is given by $\Gamma_b/\hbar$; the lifetime is given by λ_b^{-1}.

The presence of a cloud of *Z* electrons around the parent nucleus gives rise to a local (*screening*) potential, $V_e(r)$. Within the frame of the Feynman-Metropolis-Teller treatment of the equation of state [20], the electron cloud is assumed to be confined into a Wigner-Seitz (WS) cell with radius r_{WS}, within which global charge neutrality holds; r_{WS} is linked to the matter density, ρ, through the elementary relation

$$(4/3)\pi r_{WS}^3 \rho = A_w/N_A \quad (4)$$

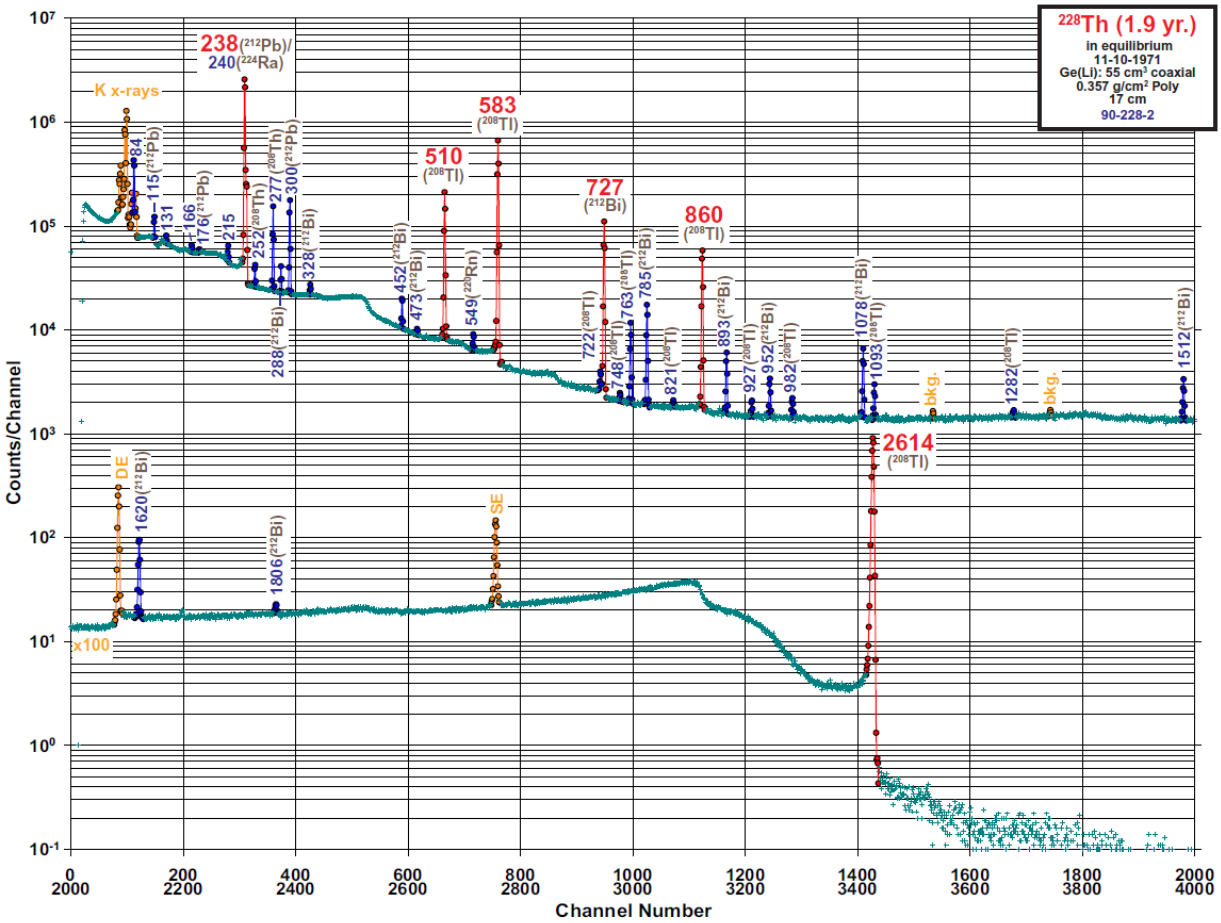

Fig. 1. γ-ray spectrum from a ^{228}Th source. Reproduced from ref. [17].

where A_w is the atomic weight and N_A is the Avogadro number; ρ is in turn fixed by the compression factor $\eta \equiv \rho/\rho_0$, where ρ_0 is a reference (*e.g.* STP) density. The interaction potential V_b and the Q-value are then modified as

$$V_b \rightarrow V(r) = V_b(r) + 2V_e(r) \qquad (5)$$
$$Q_b \rightarrow Q = Q_b + \delta E_t \qquad (6)$$

where $\delta E_t \equiv E_{t,p} - E_{t,d} - E_{t,\alpha}$, and $E_{t,p}$, $E_{t,d}$, $E_{t,\alpha}$ are the total electron binding energies inside the WS cells of the parent, daughter and α species, respectively [15]. We have utilized the generalized Thomas-Fermi (TF) model of the atom [20] to calculate $V_e(r)$ and δE_t upon compression, as described in ref. [15].

Finally, lifetime variation has been calculated through the ratio

$$\lambda/\lambda_b = (F/F_b)exp[2(G_b - G)] \qquad (7)$$

where λ, F and G refer to the decay in the electron environment, and no change in the α-preformation probability P_b has been assumed. The quantities F and G are built upon Eq. (3), as modified through the transformations in Eqs. (5,6).

α decay of ^{228}Th proceeds through several channels [17], each one with its own partial decay constant $\lambda_{b,i}$ and branching ratio B_i. Upon compression, $\lambda_b \rightarrow \lambda = \lambda_b + \delta\lambda$, where $\delta\lambda = \sum_i \delta\lambda_i$. It straightforwardly follows that

$$\frac{\delta\lambda}{\lambda_b} = \sum_i B_i \frac{\delta\lambda_i}{\lambda_{b,i}} \qquad (8)$$

Each term $\delta\lambda_i/\lambda_{b,i}$ can then be calculated according to the model described above. We have considered only the two most intense α channels in ^{228}Th decay (Table 1); they indeed

Table 1. ^{228}Th nuclear and matrix data used for calculations.

decay levels[a] (J^{π}, E [keV])	B[a] [%]	ℓ	Q[a,b] [keV]	Q_b[c] [keV]	R[d] [fm]	A_w[e] [g/mol]	ρ_0[e] [g/cm^3]
^{228}Th (0$^+$, 0) → ^{224}Ra (0$^+$, 0)	72.2	0	5520.1	5560.2	7.587	228.029	11.524
^{228}Th (0$^+$, 0) → ^{224}Ra (2$^+$, 84.373)	27.2	2					

[a] From ref. [17].
[b] Value for free atoms, in their ground state.
[c] Value derived from Eq. (6) by using the free-atom Q-value, and δE_t calculated from the binding-energy tables of Rodrigues *et al.* [21]. The energy of the i-th fed level of ^{224}Ra is then subtracted in partial width calculations.
[d] From ref. [19].
[e] From ref. [22].

account for 99.4% of the decay width. Nuclear and host-matrix parameters reported in Table 1 have been used for calculation.

In the compression domain $1 < \eta < 10$, values of $\delta\lambda/\lambda_b$ are of the order of 10^{-3}, slightly increasing with compression, which means a reduction of lifetime. In the high-pressure experiment here proposed, $\delta\lambda$ is to be measured, however, relative to the reference value, λ_0, which is found in matter in ordinary conditions ($\eta \equiv 1$). At the highest compression factor achievable in our DAC on Th ($\eta \approx 2$), we evaluated $\delta\lambda/\lambda_0 = 1.9 \times 10^{-4}$.

Experimental

A schematic of the envisaged experimental setup is shown in Fig. 2, picturing the inner part of a DAC with typical dimensions of the sample volume for reaching 1 Mbar. Indeed, we assume to work at $\eta = 2$, which corresponds to a pressure of about 1.3 Mbar for Th [18]. Prior to and after the activity measurements, the actual pressure in the DAC will have to be determined by either ruby fluorescence or X-ray diffraction at a synchrotron. Radiation effects on both techniques might need to be considered.

We plan to perform the measurement of $\delta\lambda/\lambda_0$ by using Rutherford's differential method in the version of Mazaki *et al.* [5]. Two sources, a standard (uncompressed) one (s) and a compressed one (c), which give almost the same counting rate are measured alternately with the same γ detector. Then, the difference, N, between two measured counts accumulated in a certain time interval, N_s and N_c, is assigned as the output at the time t, where we assume t to be the same for both samples. Taking into account that $N_x(t) = N_x(0)exp(-\lambda_x t)$ $(x = s, c)$, one notes that, in the limit $t\delta\lambda \ll 1$, a plot of the purposely built quantity $[N(t)/N_{c,0}]exp(\lambda_s t)$ *vs.* t is represented –apart from terms of the order $(t\delta\lambda)^2$ or smaller– by a straight-line function,

$$\frac{N(t)}{N_{c,0}} exp(\lambda_s t) = \frac{N_{s,0} - N_{c,0}}{N_{c,0}} + \delta\lambda\, t \qquad (9)$$

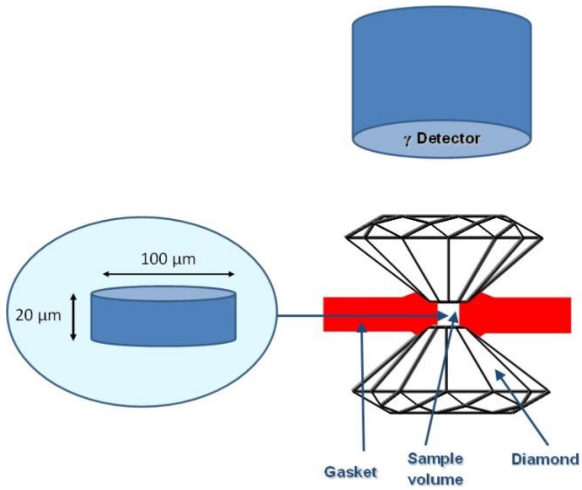

Fig. 2. Detail of the proposed experimental setup, showing the inner part of DAC, with typical sample volume size for approaching the Mbar range.

where $N_{x,0} \equiv N_x(0)$. One can finally evaluate $\delta\lambda$ as the slope of this straight line. We have performed simulations of the uncertainty affecting $\delta\lambda$ as a result of a least-squares fit on a set of *n* measurements. Curves in Fig. 3 express the fractional standard deviation, $\sigma(\delta\lambda)/\delta\lambda$, as a function of *n,* for the expected value of $\delta\lambda$ (*viz.* $\sim 2 \times 10^{-4}\,\lambda_s$) and different values of the parameters $N_{c,0}$ and Δ, the interval between two consecutive measurements (in days). As an example of a realistic scenario, if one settles for a 40%-uncertainty while working at $N_{c,0} = 4 \times 10^8$, $\Delta = 2$, a campaign of 140 measurements will be needed, lasting $n\Delta = 280$ days. Such a campaign could *e.g.* be carried out on the 2.614-MeV *γ*-rays accompanying the disintegration of ^{208}Tl, one of the decay products of ^{228}Th. With reference to Fig. 1, the corresponding spectral line is ideally suited for high-precision counting, being intense (*γ*-ray emitted in 36% of ^{228}Th decays), well isolated, and characterized by a high S/B ratio upon HPGe detection.

We finally show that, despite the low detection efficiency for such high-energy *γ*-rays (about 1% for commercial thick HPGe detectors, see *e.g.* ref. [23]), the counting rate achievable with the experimental setup of Fig. 2 is suited for our intended measurements. At the STP density indicated

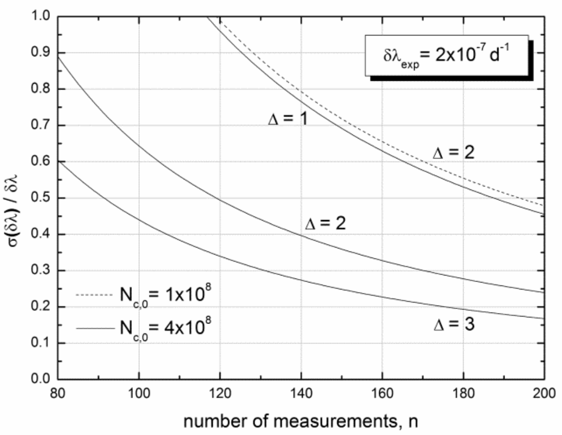

Fig. 3. Simulation of the uncertainty on $\boldsymbol{\delta\lambda}$ when retrieved from Eq. (9) by means of a least-squares fit. Curves are given as a function of the number of paired activity measurements, for relevant values of the working parameters $N_{c,0}$ and Δ.

in Table 1, the maximum amount of ^{228}Th metal that can be contained inside the DAC sample volume is about 1.8 μg, yielding a parent activity of approximately 1.5 mCi. Rescaled on the 2.614-MeV γ emission probability, it makes a total of 1.9 x 10^7 photons/s. Assuming a point source, a 10-cm^2 effective detection area at 5 cm from the source, and a detector efficiency of 1%, we expect about 6.2 x 10^3 cts/s. At this rate, the collection of 10^8 cts is achieved in about 4.5 h, which is a reasonable measurement time.

Conclusions

Experimental procedures for measuring a change in the *α*-decay lifetime of ^{228}Th as induced by static high pressure have been presented. ^{228}Th metal is a proper candidate for detecting such an effect thanks –*inter alia*– to its good compressibility, by a factor of 2 at about 1.3 Mbar. In the proposed experimental setup, the sample is compressed in a DAC, and diagnostics is based on activity-*vs*-time measurements –along a differential scheme– on suitable lines of the decay spectrum, ideally the 2.614-MeV peak from the decay of ^{208}Tl in secular equilibrium.

The small extent of the effect (our calculations predict a half-life fractional reduction of about 2 x 10^{-4} at 1.3 Mbar) and the relatively long half-life of ^{228}Th make the experiment challenging in terms of number of measurements and overall duration of the measurement campaign. Nevertheless, one has to consider that the TF model has been used to calculate atomic quantities relevant to the variation of the *α*-decay potential barrier and *Q*-value. We believe that the TF calculation provides a lower limit to the possible change in the *α*-decay lifetime compared to a more detailed quantum calculation, in which *s* electrons have a finite probability of being inside the nucleus and the electron density in the nucleus increases with pressure, as is well known from Mössbauer spectroscopy measurements

[24,25]. An effect of higher extent, up to the order of 10^{-3}, does not appear to be unlikely. The main obstacle to the feasibility of the experiment here proposed remains, however, the reduced availability of ^{228}Th in the metal form.

References

[1] E. Segré, Phys. Rev. 71 (1947) 274.
[2] R. Daudel, Rev. Sci. 85 (1947) 162.
[3] W.K. Hensley, W.A. Bassett, J.R. Huizenga, Science 181 (1973) 1164.
[4] L. Liu, C.-A. Huh, Earth Planet. Sci. Lett. 180 (2000) 163.
[5] H. Mazaki, T. Nagatomo, S. Shimizu, Phys. Rev. C 5 (1972) 1718.
[6] K. Kettner, H. Becker, F. Strieder, C. Rolfs, J. Phys. G: Nucl. Part. Phys. 32 (2006) 489.
[7] C. Rolfs, Nucl. Phys. News 16 (2006) 9.
[8] T.E. Liolios, Phys. Rev. C 68 (2003) 015804.
[9] C. Nociforo *et al.*, Phys. Scr. T150 (2012) 014028.
[10] Z. Patyk, H. Geissel, Y.A. Litvinov, A. Musumarra, C. Nociforo, Phys. Rev. C 78 (2008) 054317.
[11] H. Muir, New Sci. 36 (2006) 2574.
[12] F. Raiola *et al.*, Eur. Phys. J. A 32 (2007) 51.
[13] P. Poeml *et al.*, Phys. Rev. C 89 (2014) 024320.
[14] S. Eliezer, J. Martinez Val, M. Piera, Phys. Lett. B 672 (2009) 372.
[15] F. Belloni, Eur. Phys. J. A 52 (2016) 32.
[16] N. Nissim, F. Belloni, S. Eliezer, D. Delle Side, J.M. Martinez Val, Phys. Rev. C 94 (2016) 014601.
[17] Radiochemistry Society, Gamma-Ray Catalogue of Isotope Spectra Data, source: Idaho National Engineering & Environmental Lab., available at http://www.radiochemistry.org/periodictable/gamma_spectra.
[18] Y.K. Vohra, W.B. Holzapfel, High Press. Res. 11 (1993) 223.
[19] B. Buck, A.C. Merchant, S.M. Perez, At. Data. Nucl. Data Tables 54 (1993) 53.
[20] R.P. Feynman, N. Metropolis, E. Teller, Phys. Rev. 75 (1949) 1561.
[21] G.C. Rodrigues, P. Indelicato, J.P. Santos, P. Patté, F. Parente, At. Data Nucl. Data Tables 86 (2004) 117.
[22] C. Lavarenne, D. Mennerdhal, C. Dean, Final report - Evaluation of nuclear criticality safety data and limits for actinides in transport - C4/TMR2001/200-1, Institut de Radioprotection et de Sûreté Nucléaire, France (2004).
[23] CANBERRA Industries Inc., Broad Energy Germanium Detectors (BEGe) datasheet, available at http://www.canberra.com/products/detectors/germanium-detectors.asp.
[24] R. Ingalls, Phys Rev. 155 (1967) 157.
[25] J.A. Moyzis, H.G. Drickamer, Phys. Rev. 171 (1968) 389.