Comparison of the α-decay half-life of ^{210}Po implanted in a copper matrix at 4.2 and 293 K

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In light of recent speculations on a potentially increased speed of the radioactive decay of α-emitters embedded in a metallic matrix at very low temperature, four high-purity Cu disks deep-implanted with ^{209}Bi were prepared. The disks were irradiated in a high-flux reactor to transmute the ^{209}Bi into ^{210}Po. After the irradiation, the ^{210}Po activity was measured for all four disks. While two of the disks were stored at room temperature, the other two disks were stored in a sealed container submerged in liquid helium ($T = 4.2$ K) for approximately 200 days. After the storage time the ^{210}Po-activity measurements were repeated and the ^{210}Po half-life was calculated for each sample. No difference between the half-lives for the four samples could be detected within the uncertainties of the measurements. In other words, the results of this study show no evidence for an increase of activity for α emitters embedded in a metallic matrix at very low temperature.

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I. INTRODUCTION

It is generally accepted that the half-lives of α-decaying nuclei are constant and do not depend on their surroundings or storage conditions, e.g., temperature and chemical environment. However, in recent years, a controversial discussion started concerning the speed of radioactive α decay. Being able to change this behavior could have very high importance. In particular, accelerating the radioactive decay of long-lived α emitters could diminish one of the major challenges of nuclear waste management.

One of the key papers that was published concerning this matter appeared in 2006 by Kettner *et al.* [1]. In this paper, the authors claim that on the basis of the Debye screening model, the α-decay lifetimes for ^{210}Po and ^{226}Ra can be significantly reduced at low temperatures if the nuclei are embedded in a metal matrix. The predicted difference is of several orders of magnitude. In the case of the α decay of ^{210}Po to ^{206}Pb ($E_\alpha = 5.30$ MeV, $T_{1/2} = 138$ days, Fig. 1) the predicted half-life would be $T_{1/2} = 0.5$ days if it occurs in a metal cooled to $T = 4$ K. This corresponds to a reduction of the ^{210}Po half-life by a factor of 276.

As a final statement the authors concluded that "... one may have a cheap solution to remove transuranic waste of used-up rods of fission reactors in a time period of a few years." It is clear that this would mean a breakthrough in radioactive waste management and would receive a lot of attention in the scientific world. This matter got even more attention after an article published in the *New Scientist* in 2006 that also underpinned the applications for nuclear waste disposal [2].

One year later Raiola *et al.* [3] published experimental data for the ^{210}Po α decay that pointed in the same direction. In their experiment, the authors investigated the α decay of ^{210}Po in the metal copper at $T = 12$ K. Their finding was not as striking as their theoretical prediction; however, the authors reported that under these conditions the half-life of ^{210}Po was shorter by $6.3 \pm 1.4\%$ (as compared to the half-life at $T = 293$ K).

More recently a paper by Dong *et al.* [4] also supports this theory by experimental data. These authors used small samples of high-purity ^{209}Bi that were partly transmuted into ^{210}Po in a nuclear reactor. It was anticipated that by using this method the ^{210}Po distribution in the metal (Bi) matrix would be improved with respect to the implanted Cu samples used by Raiola *et al.* [3]. In this experiment it was found that the half-life of ^{210}Po is shortened by $24 \pm 8\%$ if embedded in high-purity metallic Bi and cooled to $T = 4.2$ K.

Despite these and other publications supporting the theory, a number of articles were published that contradict this theory. Among others, Zinner [5] published a theoretical paper in 2007 in which he demonstrates that the proposed effect [1] is unlikely from the viewpoint of standard screening models. Regarding α-decay, taking into account the electron screening effect on not only the α particle's Coulomb barrier, but also its binding energy inside the nucleus, the half-life would not significantly change. Zinner concludes that even at temperatures of the order of a few Kelvin, any change should be negligible and should actually point towards an enhancement rather than a reduction of the half-life.

Wauters *et al.* [6] investigated the α decay of ^{221}Fr embedded in Si at $T = 4$ K as well as embedded in Au at $T = 4$ K and $T \approx 20$ mK. The authors report that within $\pm 0.1\%$ they did not find any difference in half-life for the different conditions. Furthermore, the authors argue that if there was any such effect it should have been observed previously in low-temperature nuclear orientation (LTNO) experiments.

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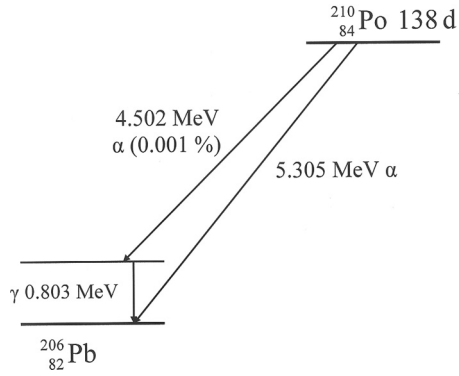

FIG. 1. Decay scheme of ^{210}Po.

However, these LTNO experiments on α-decaying isotopes implanted in Fe showed no difference in half-life between room temperature and very low temperatures [7]. Several more experimental studies on the α decay of ^{253}Es, ^{224}Rn, ^{225}Ra, and ^{227}Ac did also not evidence any sign of a change of half-life [8,9]. Olaizola *et al.* [10] studied the dependence of the half-life of ^{221}Fr on the implantation environment (Si or Au) and found that the difference in half-life does not follow a systematic trend and is well below 1%.

Regarding the half-life of ^{210}Po, Pierre *et al.* [11] performed an experiment where ^{210}Po was electrodeposited on a silver plate. The resulting material was then covered by a Ag foil, heated, and pressed, to completely encase the ^{210}Po by Ag. Here too, the authors did not detect any significant change (neither longer nor shorter) of the half-life of ^{210}Po.

To contribute to this important debate about the half-life of ^{210}Po in a metal matrix at low temperature, we report in this study the results of an ad-hoc experiment on ^{210}Po embedded in an ultrapure Cu matrix and stored at liquid He temperature, i.e., $T = 4.2$ K. The experiment was designed to tackle as much as possible the ambiguities and shortcomings of previous studies reported on this topic [3,11].

II. EXPERIMENT

A. Experiment design

The design of the present experiment is based on that of Raiola *et al.* [3]. By repeating their experiment as an independent group we intended to verify their results. We applied, however, several changes to the original setup that we think address some of the ambiguities that could have arisen.

(i) The ^{210}Po activity was measured outside the cryostat, i.e., at room temperature. This avoids any temperature-dependent detector gain corrections and other unwanted low-temperature effects on the samples (e.g., thermal contraction).
(ii) The α activities of all samples were measured in the same dedicated spectrometer chamber to eliminate any geometry effects.
(iii) The storage temperature was decreased down to 4.2 K to magnify any potential effect.
(iv) To maximize the activity measurement sensitivity with respect to the variation of lifetime, the samples were held at cryogenic temperature for one entire lifetime of ^{210}Po, i.e., 200 days.
(v) The activation conditions in the reactor were set to achieve a higher ^{210}Po activity in our samples compared to the ones used in Ref. [3], improving the signal-to-background ratio and the counting statistics.
(vi) The samples, originally implanted with ^{209}Bi as in Ref. [3], were prepared to exhibit a broader depth profile and a deeper localization of ^{210}Po after transmutation. A broad depth profile helps to increase the sample activity while keeping doping at less than 1 at. % and reducing the concentration of implantation-induced lattice defects. Deeper implantation rules out any possible effect of surface oxidation and contamination, which would locally alter the metallic properties of the host matrix [3,4,6,11].

B. Sample preparation

Four Cu disks (99.9871 wt. %, ø = 10 mm, 0.5 mm in thickness) were implanted at the Tandem Van-de-Graaff accelerator of the Max-Planck-Institut für Kernphysik, Heidelberg. A polyenergetic ^{209}Bi ion beam was used (15–60 MeV; ion charge states between +1 and +9), which spanned implantation doses in the order of 10^{15}–10^{16} atoms (depending on the sample) over the range 1–4 μm beneath the disk surface, resulting in a Bi/Cu concentration lower than 1 at. %, as calculated by means of the code SRIM-2010 [12].

To investigating any possible role played by oxidation, thin Cu foils were also prepared, bearing ^{210}Po only within a narrow surface layer (below 100 nm). These samples, however, did not survive neutron-irradiation damage and were unusable for measurements. Four Cu thin foils (99.8668 wt.%, ø = 10 mm, 6 μm in thickness) were shallow-implanted (within 100 nm from the surface) with ^{209}Bi via a laser-ablation ion source [13] at the Laboratory of Applied Electronics, Department of Physics, University of Salento. The implanted/codeposited dose was about 10^{15} atoms, resulting in a Bi concentration of about 1 at. %.

C. Sample irradiation

The eight Cu disks were irradiated at the High Flux Reactor (HFR) at Petten (The Netherlands) to transmute the ^{209}Bi implanted in the disks to ^{210}Po. The irradiation was performed in an "in-core position" (E3), the neutron flux in that position is given in Table I.

The disks were placed in a specially manufactured disk holder made of titanium, which was then double-encapsulated in Ti irradiation capsules. To reach the activation required, the disks were irradiated for 29 days. An estimate of the activity reached was calculated with the code FISPACT [14]. To assess the temperature of the disks during irradiation, the finite element code ANSYS was used [15].

The 0.5-mm-thick disks reached a maximum calculated ^{210}Po activity of 5.34×10^4 Bq 15 days after the end of the irradiation. The 6-μm-thick disks reached a maximum calculated ^{210}Po activity of 1.76×10^4 Bq, also 15 days after the end of the irradiation.

TABLE I. Neutron fluence rates at different energies in reactor core position E3.

	Average Fluence rate in E3 (10^{18} m^{-2} s^{-1})	Neutron Energy (E)
	1.09	$E < 0.058$ eV
Thermal	0.919	0.058 eV $< E <$ 0.248 eV
	0.237	0.248 eV $< E <$ 0.625 eV
⇓	0.436	0.625 eV $< E <$ 4 eV
	1.76	4 eV $< E <$ 5.53 keV
Fast	2.32	5.53 keV $< E <$ 0.82 MeV
	1.89	$E >$ 0.82 MeV

After extraction from the irradiation capsule, all the samples were cleaned in an ultrasonic bath. However, in the case of the 6-μm-thick samples, ultrasonic cleaning resulted in their disintegration, probably owing to embrittlement induced by fast-neutron irradiation.

D. Liquid-helium storage

The activity of the four disks was measured immediately before and after a storage period of ≈200 days, i.e., one ^{210}Po lifetime. Two disks were stored in liquid helium ($T =$ 4.2 K), while the other two were kept at room temperature ($T =$ 293 K).

For the long-term and safe storage of the samples at liquid-helium temperature, but also to ensure their proper thermalization, a special capsule (inner containment) and a container (outer containment) were designed (Fig. 2). The samples were placed into the capsule; the capsule was then inserted into the container. Both the container and the capsule were filled with helium exchange gas. The capsule was sealed with an indium O-ring; the container was sealed with Stycast glue (Stycast 2850 FT) to prevent any intrusion of liquid helium into the container. Then the container was mounted onto a long rod that was inserted into a liquid-helium tank (ca. 50 l in volume) and submerged in liquid helium. The liquid helium level was kept at all times above the metallic container ensuring that the samples were maintained at $T = 4.2$ K (by thermal conduction of the container immersed in the liquid helium bath) during the whole duration of storage. Note that the size of the liquid-helium tank (and hence the mass of liquid helium) was much larger than the size of the sample container (and hence the mass of the samples) canceling any thermal gradient between the liquid helium bath and the disks containing only a diminutive amount (in the range of nanograms) of ^{210}Po despite its relatively huge self-heating power (141 W/g). At the end of the storage, the glued container was warmed up to room temperature and the samples were sent for activity measurements.

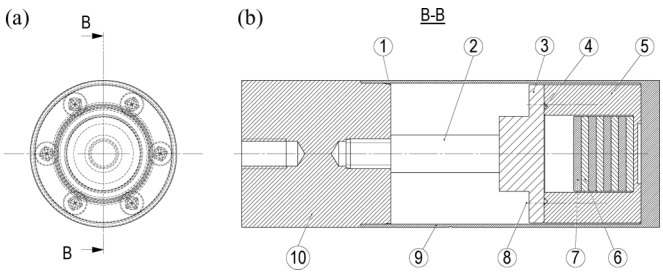

FIG. 2. Storage of the samples at low temperature. (a) Top view of the internal capsule (sealed with indium wire and screwed). (b) Sectional view (B-B) of the whole container. (1) Stycast glue seal, (2) plastic rod holding the internal capsule in place, (3) cap of the internal capsule, (4) indium O-ring, (5) body of the internal capsule, (6) samples (four are represented here but only two 0.5-mm disks were effectively loaded, see text), (7) aluminium disks separating the samples, (8) screw, (9) body of the external container, and (10) head of the external container.

E. α-activity measurements

The α-activity measurements were carried out on an Ortec Spectrum Master 920-8 equipped with an Ortec EG&G Soloist chamber and an ion-implanted Si detector (Ametek) with a surface of 900 mm^2. The chamber was equipped with a special sample holder to ensure precise and reproducible positioning of the disks. The distance between sample and detector was 21 mm. Energy and efficiency calibration of the detector were accomplished by using a certified α source (AMR 43 manufactured by Amersham).

The chamber was dedicated to the presented measurements; i.e., no other samples were introduced into the chamber during the whole period of the project. The background of the chamber was measured both before and after the measurement of a sample. The efficiency of the detector was determined using the AMR 43 source both at the start and at the end of the experiment and was found to be 0.0593(9) and 0.0595(9), respectively.

The uncertainty on the results was calculated taking into account the given uncertainties on the AMR 43 reference material, the statistical counting error on the efficiency measurements, and the statistical counting error on the α-activity measurements of the samples. All uncertainties are given as 1σ.

The counting time of the samples was adjusted to achieve a total of 10^6 counts and thus a statistical error of 0.1%. The results were corrected for the decay of the ^{210}Po during the measurement using the equation

$$R_{\mathrm{corr}} = \frac{R\frac{t_{\mathrm{m}}\ln 2}{T_{1/2}}}{1-\exp\left(-\frac{t_{\mathrm{m}}\ln 2}{T_{1/2}}\right)}, \tag{1}$$

where R_{corr} is the instrument reading corrected to the beginning of the individual measurement, R the background-corrected instrument reading before decay correction, t_{m} the time of the individual measurement, and $T_{1/2}$ the half-life of the measured nuclide. The value 138.3763(17) d was used for the half-life of ^{210}Po [16].

The half-life of the measured samples was calculated using the equation

$$T_{1/2} = \frac{-\ln 2t}{\ln\left(\frac{R_{\mathrm{corr}}}{R_{\mathrm{corr},0}}\right)}, \tag{2}$$

where $T_{1/2}$ is the calculated half-life, $R_{\mathrm{corr},0}$ and R_{corr} are the initial activity intensity and the final activity intensity,

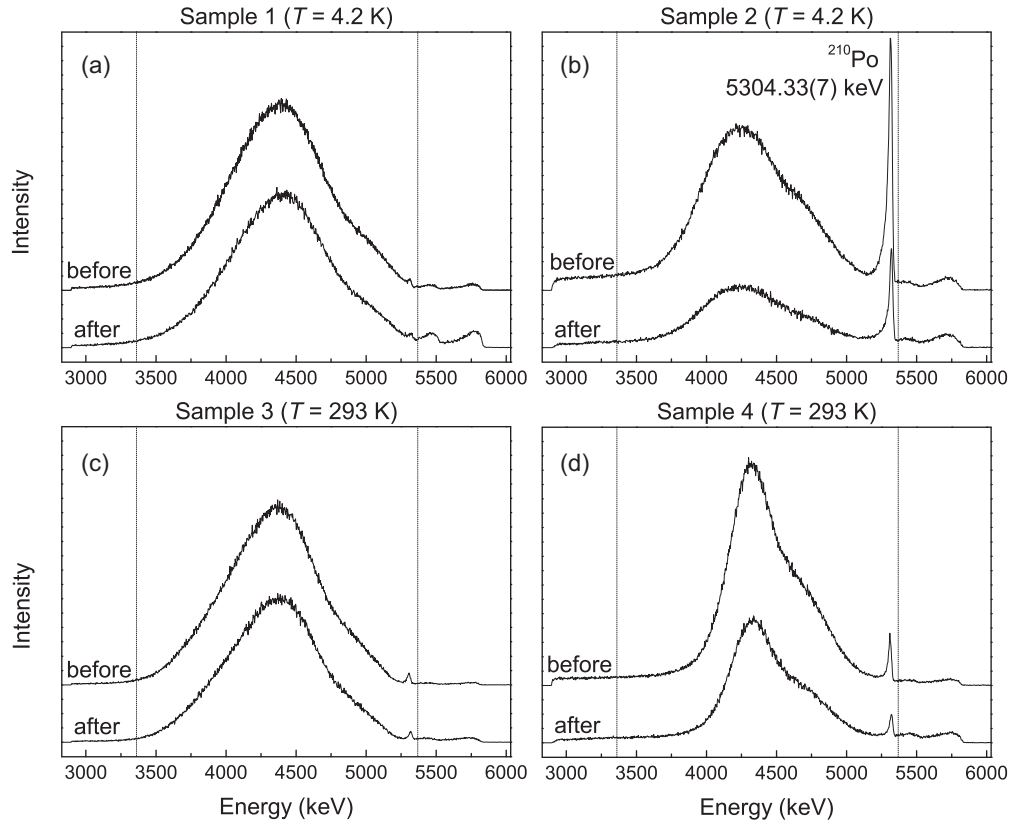

FIG. 3. α-spectra of the four samples before and after storage. The peak of the α decay of ^{210}Po (5304.33 keV) is clearly visible in panel (b) (sample 2). The two vertical lines in each graph delimit the region of interest that was chosen to evaluate the spectra.

respectively, and t is the time elapsed between the beginnings of the measurements of $R_{\mathrm{corr},0}$ and R_{corr}.

III. RESULTS AND DISCUSSION

Figure 3 shows the α spectra of the four disks before and after storage. The two vertical dotted lines in each graph depict the limits of the region of interest that was used to calculate the activities of the samples. From the spectra it is evident that the Bi implantation into the samples did not produce exactly the same result for each specimen. The shapes of the spectra are different for each sample, as is the intensity of the ^{210}Po peak at 5304.33 keV. This leads to the assumption that for each sample a varying amount of ^{210}Po is located near its surface. The majority of the α intensity, however, comes from the broad peak located at a lower energy ($\approx$4300 keV) compared to the ^{210}Po peak. This broad peak is the result of α particles from deeply implanted ^{210}Po that suffer a certain energy loss while travelling to the sample surface. This proves that the gross of ^{209}Bi was indeed deeply implanted into the Cu (1–4 μm) and hence properly surrounded by Cu metal atoms. Note that the difference in the shape of the α-spectrum does not affect the results, because for the method of half-life determination used no comparison between individual samples is necessary. In this context it is also noteworthy that no indication for ^{210}Po diffusion to the surface of the samples was found in this experiment.

A summary of the measured α activities and half-lives is given in Table II. The α activities of the four samples range from 54 to 479 Bq before sample storage and from 16 to 139 Bq after storage.

The determined half-lives of ^{210}Po range from 139.21 ± 2.90 to 143.29 ± 2.98 d. Figure 4 shows a plot of the four determined half-lives along with the reference value. All determined half-lives are longer than the reference value of 138.3763(17) d [16]; however, all five values lie within their 2σ uncertainties. We hence conclude that within the measurement uncertainty there is no evidence for a difference in half-lives

TABLE II. Summary of the data acquired in this experiment. The uncertainties are given as 1σ.

		$T = 4.2$ K		$T = 293$ K		Average
		Sample 1	Sample 2	Sample 3	Sample 4	
Activity (Bq)	Before storage	479	54	434	71	
	After storage	139	16	132	22	
$T_{1/2}$ (d)		140.2 ± 2.9	139.2 ± 2.9	139.6 ± 2.9	143.3 ± 3.0	140.6 ± 1.5

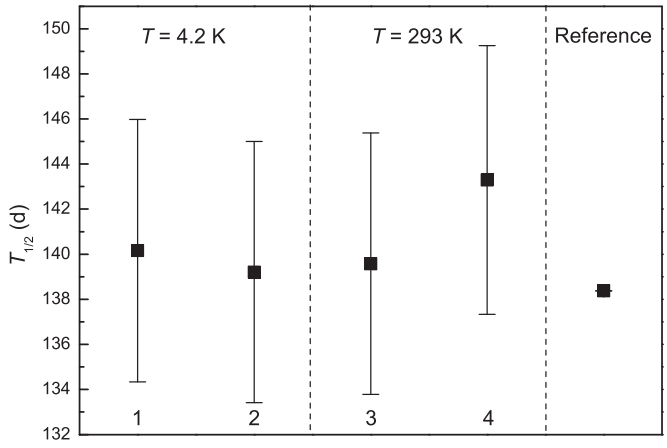

FIG. 4. Plot of the four determined half-lives along with the reference value (error bars represent 2σ). All four half-lives are longer than the one given in the reference.

between the samples stored at $T = 4.2$ K and those stored at $T = 293$ K. In particular, no decrease in half-life could be detected.

IV. CONCLUSIONS

Four Cu disks were successfully implanted with ^{209}Bi and subsequently irradiated to transmute the ^{209}Bi to ^{210}Po. The ^{210}Po α activities were measured before and after storage of two disks at room temperature and two disks in liquid He. All four measured half-lives were found to be slightly higher than the reference value of 138.3763(17) d [16].

Within the measurement uncertainty, no decrease of the half-life of ^{210}Po was detected in this study. A value of $T_{1/2} = 129.66$ d as reported in Ref. [3] or a value of $T_{1/2} = 105.2$ d as reported in Ref. [4] could not be reproduced.

This result is in-line with early and recent calculations. The tunneling probability of the α particle through the Coulomb potential barrier is one of the factors determining the α-decay lifetime. The possibility that this quantity could be modified by a cloud of quasifree metallic electrons around the nucleus, partially screening the barrier, was first considered by Salpeter in 1954 [17]. Early calculations, however, showed that the effect was negligible [18]. After the claims made in Ref. [1], the theoretical problem was revisited. Zinner [5] showed that the tunneling probability is not substantially affected by the electron screening if the lowering of the tunneling barrier is accompanied by a compensating change in the binding energy of the α particle. In the framework of the Debye plasma model, Eliezer *et al.* [19] calculated a decrease of about 1% or less (much smaller than the one predicted in Ref. [1]) of the decay lifetime for ^{210}Po embedded in a Pd matrix at $T = 4$ K.

To conclude, our results demonstrate that no dramatic lifetime changes occur when the α emitter is dispersed in a metallic matrix cooled at liquid-helium temperatures.

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