Eur. Phys. J. A **32**, 31–34 (2007)
DOI 10.1140/epja/i2007-10011-9

THE EUROPEAN PHYSICAL JOURNAL A

Regular Article – Nuclear Structure and Reactions

Alpha-decay half-life of ^{221}Fr in different environments

H.B. Jeppesen[1,a], J. Byskov-Nielsen[1,2], P. Wright[1,3], J.G. Correia[1], L.M. Fraile[1], H.O.U. Fynbo[2], K. Johnston[1], and K. Riisager[1]

[1] ISOLDE, PH Department, CERN, CH-1211 Genéve 23, Switzerland
[2] Institut for Fysik og Astronomi, Aarhus Universitet, DK–8000 Aarhus C, Denmark
[3] Fundamental Fysik, Chalmers Tekniska Högskola, S-412 96 Göteborg, Sweden

Received: 30 January 2007 / Revised: 5 March 2007
Published online: 3 April 2007 –
Communicated by J. Äystö

Abstract. A possible dependence of the alpha decay process on the solid-state environment of the decaying nucleus has been investigated by measuring the alpha-decay half-life of ^{221}Fr at room temperature in different materials (Au, W, Si and polyethylene). The change in half-life between the insulator Si and the metals Au/W is found to be 0.30(17)% / 0.42(21)%, respectivly. Furthermore an improved value of 4.79(2) min was found for the absolute half-life of ^{221}Fr.

PACS. 23.60.+e Alpha decay – 21.10.Tg Lifetimes – 27.90.+b $220 \leq A$

1 Introduction

In the first decades after radioactivity was discovered it was established that nuclear decay rates to a large degree are independent of outside perturbations [1,2] and they are therefore in essentially all textbooks considered an intrinsic property of the decaying nuclear state. Although this is a good approximation, we know today that atomic effects can change electron capture beta decay and internal-conversion decay rates by up to roughly one percent [1–3] and the influence of atomic electrons in beta decays is also well established. Alpha decay rates may also vary, according to a recent suggestion by C. Rolfs and collaborators [4,5], depending on the density of quasi-free electrons in the environment around the decaying nucleus. Such an effect has been reported for weak decays in two new experiments where the half-lives of ^{22}Na (β^+ decay) [6] and ^{7}Be (electron capture) [7] were seen to vary by 1.2(2)% and 0.9(2)%/0.7(2)% (in Pd/In) when samples were cooled down to 12 K. Furthermore a possible increase in the half-life of ^{7}Be at room temperature was recently reported [8]. A similar systematic effect that increases at lower temperatures has been observed for many metals in low-energy nuclear reactions [4,5,9], but is not yet understood theoretically. Simple estimates [4,5] give changes in alpha decay rates that even at room temperature might be a factor of two or more. We report here on a search for this effect using the alpha-decaying nucleus ^{221}Fr.

[a] e-mail: henjep@phys.au.dk

2 Experimental setup

The experiment was performed at the CERN/ISOLDE PS-Booster facility [10] where a 1.4 GeV proton beam impinged on a UC_x target producing a wide variety of nuclei. The produced nuclei then diffused out of the target and into a tungsten surface ion source. The ions were extracted with 60 kV, the ^{221}Fr ions were selected in the ISOLDE General Purpose Separator and subsequently led to the experimental setup.

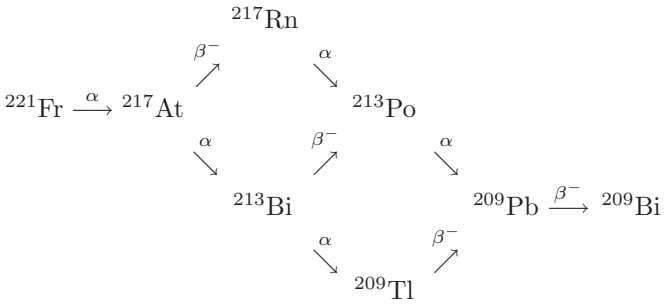
The detector setup consisted of a turnable target ladder and a surface barrier Si detector of 500 μm thickness and 300 mm^2 active area. The detector covered a solid angle of about 2%. Four samples were mounted on the target ladder (W, Au, Si and polyethylene) two on either side. These metals were chosen due to large observed electron screening in d-d reactions with deuterons implanted into these metals [9], and due to their inherent resistance to form oxide layers. A ^{221}Fr sample was typically collected for some seconds and then turned 180° to face the Si detector. Data taking started about one minute after the collection ended and the decay of each sample was followed for about 2 hours (about 20 half-lives). The procedure of collecting samples in the opposite geometry to that in which the detection took place was followed to suppress events from longer-lived contamination in previously collected samples. For each sample a total of four collections were made except for the Au sample where the number was five. The yield of ^{221}Fr decreased towards the end of the experiment which increased the collection time to a few minutes.

The implantation and subsequent measurement was performed in a clean environment with a vacuum of about 10^{-6} mbar.

3 Data analysis

3.1 Recorded data

The decay chain of ^{221}Fr involves several daughter nuclei connected in the network shown below:



The half-life of ^{221}Fr, 4.9(2) min, is much longer than the one of the daughter ^{217}At, 32.3(4) ms, and the time distribution recorded for the alpha particle from ^{217}At will therefore also display the ^{221}Fr half-life. In a similar way the time distribution recorded for the alpha particle from ^{213}Po will show the half-life of ^{213}Bi, 45.59(6) min, rather than its own half-life, 4.2(8) μs. All quoted half-lives are taken from [11].

An example of an energy spectrum is shown in fig. 1 with the origin of the peaks identified by labels. In addition to the expected daughters from ^{221}Fr decay we also see contributions from contaminants in the beam (^{221}Ra and in the longer collections at the end of the run also Rn isotopes and ^{222}Fr). For the strongest of the contaminant peaks their identity was confirmed with half-life analyses. The amount of these contaminants varied between samples. For the ^{213}Po peak the high-energy tail is caused by summing with the preceding beta particle from decay of ^{213}Bi.

For the two metals considered here, it is useful to have an appreciation for the implantation depths of the Fr ions and to take into account any influence from oxide layers on the surface. Using the SRIM program one can gain an insight into the depth: for Au and W one obtains a mean implantation depth value of $\simeq$ 80 Å in both cases with a 40 Å distribution. For the Au sample no oxide layer is expected to form, whilst for W one would expect an oxide of about 20–30 Å on the surface, for the conditions under which the experiments were conducted. The Au sample can therefore be considered as a "clean" system with the ions incorporated in the bulk. From SRIM calculations less than 5% of the Fr ions is seen to stop in the WO_2 layer thus producing a source which is also relatively clean. From numerous solid-state experiments it has been shown that ions implanted in this manner will be in a metallic environment, see also the discussion in [12].

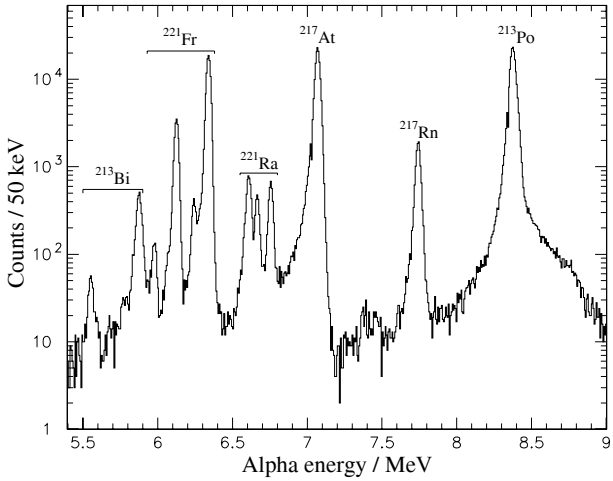


Fig. 1. An example of α energy spectrum (W samples).

One important possible source of systematic error in the present experiment is diffusion. If a sizeable fraction of the sample diffuses out of the target this would yield an effective shorter half-life which could be misinterpreted as a screening effect. We have earlier observed significant diffusion out of polyethylene targets implanted with noble gases at ISOLDE [13], and have therefore included this material in our list of samples. The observed half-life in polyethylene was in perfect agreement with the ones measured for Si, but clear evidence of diffusion was observed in the energy spectrum where the main ^{221}Fr peak and its side peak were not separable.

The recoiling daughter nuclei will have kinetic energies around 130 keV and will therefore have a larger range in the samples than the implanted 60 keV ions. The depth distribution of the daughter nuclei will thereby be smeared out and a fraction will have sufficient energy to leave the sample again. We do see in all spectra a clear broadening of the alpha peak from ^{217}At compared to the ones from ^{221}Fr, the enhanced low-energy tail is clearly visible in fig. 1. Note that this implies that the diffusion of Fr in our samples (expect for polyethylene) is constrained to be clearly smaller than the recoil range. From this we conclude that the effect of diffusion on our extracted half-lives is below our uncertainty.

To monitor deadtime of the data aquisition a 50 Hz pulser was fed into the data stream. By comparing the number of missing pulser events with the instantaneous count rate the deadtime was extracted to be 55 μs. This is in very good agreement with a goodness-of-fit test (Anderson-Darling) performed on the quality of the half-life fits for different values of the deadtime. The deadtime is to a very good approximation constant for all events since all data events had the same structure. To correct for deadtime in the half-life fits we multiplied the fit function by the factor $(1 - N_{tot}t_{dead})$, where N_{tot} is the instantaneous number of event per second and t_{dead} is the deadtime. In the most extreme cases the correction factor was up to a few %. In this respect, our results are similar to the ones reported recently [8] on ^{7}Be.

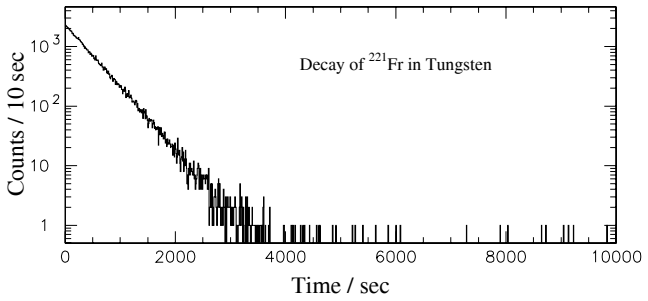


Fig. 2. An example of the time spectrum from the decay of ^{221}Fr in W samples.

Table 1. Half-life in min of ^{221}Fr implanted in different host materials evaluated from two different gates.

Sample	^{221}Fr gate	^{217}At gate
Au	4.789(6)	4.779(5)
W	4.782(8)	4.775(6)
Si	4.802(7)	4.795(6)
CH_2	4.804(8)	4.791(7)

3.2 Results

The half-life was determined from the time spectrum of events falling within a narrow energy gate of the main ^{221}Fr peak (6.275–6.400 MeV, see example in fig. 2). The different collections were analyzed independently, the final weigthed averages for each sample are shown in table 1. (For both Au and W samples the scatter among results from the collections was larger than statistically expected.) The table also shows the results from events within a small gate around the ^{217}At peak; the half-lives derived here are consistent with the ones from ^{221}Fr but slightly smaller. We shall in the following only use the ^{221}Fr data to avoid possible complications arising from the changes in source extent due to the nuclear recoils.

The absolute time calibration was done by adjusting the fitted half-life of the decay of ^{213}Bi to the literature value of 45.59(6) min [11]. This procedure was performed with data from Si samples in order to avoid possible shifts from the metal samples. From this absolute time calibration an improved value for the half-life of ^{221}Fr of 4.79(2) min could be extracted which is a clear improvement from 4.9(2) min [11].

The half-lives measured in the metallic samples are systematicly shorter than the ones measured in silicon, but with a difference of 0.30(17)% and 0.42(21)% for Au and W the effect is small. The statistical significance of the present data set is not sufficient to clearly show an effect, but we can safely place an upper limit of 1% on the relative half-life difference.

4 Discussion

The results we obtain are clearly smaller than the ones suggested by application of the phenomenological Debye model [4], where the key ingredient is the screening from the quasi-free metallic electrons that is assumed to be given by the estimate from the Debye plasma model. We shall in this section look in more detail at the influence of screening on alpha decays, but would like to point out first that the Debye model may turn out to miss important physics ingredients; a quantitative theoretical explanation of the enhancements observed in low-energy nuclear reactions is still missing in spite of several attempts [14,15].

It is useful to recall the discussion of atomic effects on alpha particle decays, *i.e.* the difference between the decay of a bare nucleus and of a nucleus imbedded in a neutral atom. A good basic discussion can be found in [16,17]. Considering the difference in potential energy between a neutral atom and a bare nucleus due to the potential of the atomic electrons on the nucleus, one would derive a difference in alpha particle energy in the two cases of about 34 keV for Fr. Since the alpha particles are significantly slower than the inner-shell electrons, the latter adapt adiabatically to the alpha decay and the probability of seeing shake-up and shake-off from the inner shells is as a consequence very small. The average energy going into atomic excitation is therefore also small, estimates give rougly 250 eV. So the main effect of "dressing an atom with electrons" will be that the energy of the outgoing alpha particle is reduced by the potential energy difference. This is in a way obvious: in the atom the nucleus is placed at a lower starting potential, so the energy at infinity of the emitted alpha will be decreased. The average atomic excitation energy is of course a value that could change when going to a nucleus imbedded in a solid. However, this is unlikely to be relevant for the decay probability since the shake processes happen at a distance beyond the outer turning point in the tunneling process.

Now, the final energy of the alpha particle, its kinetic energy when it is at infinite distance from the nucleus, is actually not the relevant energy when calculating the barrier penetration probability. If the inner part of the atom is moved up or down by a constant potential energy this does not change the barrier penetration —only changes in the region between the inner and outer classical turning point are relevant. In [1] the example of ^{226}Ra is given, the total potential difference from the electron cloud between the inner and outer classical turning points is given as 124 eV. One is normally only able to change very little on the electronic cloud (*e.g.*, by inserting the atom in different chemical compounds) and the measurable chemical shifts will be negligible. A more accurate expression for the effect of a change of the atomic potential in the barrier region is given in [18].

Returning now to the Debye model, the Debye radius for the d+d reactions is about one tenth Bohr radius at room temperature, *i.e.* 5000 fm (scaling as the square root of the temperature). If this is scaled with the nuclear charge one reaches distances comparable with outer turning points which are at 30–40 fm. Since the screening electrons will be even faster than the inner-shell electrons they should also respond adiabatically to the outgoing alpha particle. The energy of the alpha particle will therefore be

changed even more than for the neutral atom case[1]. Concerning the decay probability, if the screening potential is still approximately constant in the "tunneling region", no changes are expected. If one can feel the rise in the screening potential before the outer turning point is reached, the tunneling barrier will be larger and the half-life should become longer. The formulas given in [18] should in principle allow for quantitative estimates for given positions of the screening electrons.

If there is significant extra screening in metals, the standard atomic structure is perturbed. The screening electrons decrease the atomic potential, so the Fr/At electrons would become less bound and move outwards. One can thus not simply add the contribution from the Debye model to the ones from the atom case. This is mainly relevant for the energy scale, where the atomic contribution to the change in final outgoing alpha particle energy will become smaller. However, the overall effect of extra screening will still be an increased energy shift.

We note that an independent critique of the application of the Debye model to alpha decay has been published recently [19].

5 Conclusion

We have looked for an environment dependence of alpha decay half-lives by directly measuring the half-life of ^{221}Fr implanted in Au, W, Si and polyethylene. The two metal samples show a slightly faster decay than the insulators, but all samples agree within two standard deviations and an improved half-life of ^{221}Fr of 4.79(2) min has been found from the Si samples. The change in half-life is clearly less than 1% at room temperature, which is considerably below the simple estimates from the phenomenological Debye model. The absolute energy of the alpha particles would also change if the screening around the alpha-decaying nucleus is changed significantly in metals compared to insulators, from our data we could only set a crude upper limit on such an energy change. For future searches of this effect it would be valuable also to look at samples at different temperatures.

This work was supported by the ISOLDE Collaboration. We are grateful to Prof. C. Rolfs for drawing our attention to the possible half-life variability and to Prof. H. Haas for information on Fr kinetics in different materials.

References

1. G.T. Emery, Annu. Rev. Nucl. Sci. **22**, 165 (1972).
2. H.-P. Hahn, H.-J. Born, J.I. Kim, Radiochim. Acta **23**, 23 (1976).
3. M.S. Freedman, Annu. Rev. Nucl. Sci. **24**, 209 (1974).
4. F. Raiola *et al.*, J. Phys. G **31**, 1141 (2005).
5. K.U. Kettner *et al.*, J. Phys. G **32**, 489 (2006).
6. B. Limata *et al.*, Eur. Phys. J. A **28**, 251 (2006).
7. B. Wang *et al.*, Eur. Phys. J. A **28**, 375 (2006).
8. Y. Nir-El *et al.*, Phys. Rev. C **75**, 012801(R) (2007).
9. F. Raiola *et al.*, Eur. Phys. J. A **19**, 283 (2004).
10. E. Kugler, Hyperfine Interact. **129**, 23 (2000).
11. G. Audi, O. Bersillon, J. Blachot, A.W. Wapstra, Nucl. Phys. A **729**, 3 (2003).
12. N.J. Stone *et al.*, Preprint, arXiv:nucl-ex/0611041.
13. U.C. Bergmann *et al.*, Nucl. Phys. A **714**, 21 (2003).
14. F. Raiola *et al.*, Phys. Lett. B **547**, 193 (2002).
15. K. Czerski *et al.*, Europhys. Lett. **68**, 363 (2004).
16. R.G. Thomas, Prog. Theor. Phys. **12**, 253 (1954).
17. I. Perlman, J.O. Rasmussen, Handb. Phys. **XLII**, Nucl. React. III, pp. 151-153 (1957).
18. W. Rubinson, M.L. Perlman, Phys. Lett. **40**, 352 (1972).
19. N.T. Zinner, Nucl. Phys. A **781**, 81 (2007).

[1] We have looked for a systematic sample-dependent shift in the alpha particle energies in our data, but did not find any. Due to a slow overall drift in the electronic amplification we can put a conservative upper limit on such a shift of 10 keV, only.