Proposal to the ISOLDE and NTOF Committee

Investigation of alpha-decay rates of $^{221}$Fr, $^{224}$Ra and $^{226}$Ra in different environments.


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Abstract

It has recently been suggested, and indicated experimentally, that alpha decay half-lives are modified by solid state effects in the surrounding environment. We propose here to measure with high accuracy the alpha-decay half-life of $^{221}$Fr, $^{224}$Ra and $^{226}$Ra in insulators and metals. Furthermore we plan to investigate the temperature dependency of the half-life in these materials (room temperature, 4 K and 10 mK).

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1 Motivation

A number of experiments performed in the first decades after radioactivity was discovered showed that the nuclear decay rates cannot be changed by outside perturbations [1, 2]. This conclusion is still generally accepted today and the decay rate is considered an intrinsic property of the decaying nuclear state. However, more detailed investigations have shown that atomic structure in several instances affects nuclear events, the most well-known example being the cases of electron capture beta decay and internal conversion where decay rates can vary by up to roughly one percent [1, 2, 3]. This is not surprising since both processes depend directly on the density of atomic electrons surrounding the nucleus. An elegant series of experiments at the ESR storage ring at GSI has during the last decades shown how also beta decay process will vary as the charge state of the atom is changed [4]. The question we address in this proposal is whether also alpha decay halflives may depend on the nuclear surroundings.

That this question should be answered in the affirmative is suggested in several recent papers [5, 6] based on the observation that the rate of nuclear reactions at very low energy depend on whether they take place in an insulator (or a gas) or in a metal. Screening effects are very important for nuclear reactions in the keV energy range and a series of experiments by several independent groups have all observed rate differences that are caused by different amounts of screening in insulators and metals [7, 8, 9]. A quantitative theoretical understanding of the effect is still lacking in spite of several attempts [10, 11], but the Debye plasma model has been suggested by the group of Rolfs as a parametrization of existing data [5, 6]. The extra screening is here attributed to the valence electrons of the metal and the model also reproduces an observed significant temperature dependence of the effect [5]. Two very recent experiments [12, 13] reported changes of the halflives of $^{22}\text{Na}$ ($\beta^+$ decay) and $^{7}\text{Be}$ (electron capture) on the one percent level for samples implanted in metals cooled to 12 K, in qualitative agreement with the trend expected from the Debye model. A 10 % shorter halflife is in a similar way obtained for the $^{210}\text{Po}$ alpha decay [14].

Screening effects in alpha decays would, in analogy to the effects seen when going from a bare nucleus to an atom [15, 16], be expected to change the observed energy of the alpha-particle much more than the decay rate [17]. However, the lack of a solid explanation for the effects observed in reactions and weak decays and the substantial consequences (scientifically as well as in applications [6]) a change in alpha-decay halflife would have, necessitates a thorough investigation of this possibility.

We propose two different lines of investigation. In the first method we investigate long lived nuclei by comparing the activity of a sample at different temperatures. The second method uses short lived isotopes to measure directly the halflives in different materials and at different temperatures.

2 Activity measurement of $^{226}\text{Ra}$

The first approach measures activities of long lived samples. This is the method used in [12, 13, 14]. We suggest here to use $^{226}\text{Ra}$ with a halflife of 1600 years. For this isotope an effect as large as a factor of 1000 has been proposed [6].

ISOLDE is one of the few places where pure and intense sources of $^{226}\text{Ra}$ are available. We will need to collect $10^{12}$ atoms in each of four samples, Cu, Au, Pd and Si, at the ISOLDE HV platform. A source strength of 10 Bq ($10^{12}$ atoms) is needed to be able to perform measurement with a statistical precision better than 1% within 24 hours. A much stronger source would only give rise to deadtime and provide us with a less pure
environment.

This required source of $10^{12}$ atoms should be possible since the ISOLDE data base quotes SC yields of $1.1 \times 10^8/\mu$C from 13 g/cm² UC and $1.1 \times 10^8/\mu$C from 55 g/cm² ThC (from decay of $^{226}$Fr) which corresponds to collection times between 3 hours and 20 minutes. The collected samples will each have activities on the order of 10 Bq and will therefore pose no radiation hazard.

The activity measurements will be performed down to a temperature of 1 K at Isotopenlabor at Ruhr-Universität, Bochum, Germany which have expertise in measuring activities. In this case the gamma-rays from $^{226}$Ra are used to monitor to activity.

3 halflife measurements of $^{221}$Fr and $^{224}$Ra

In the second approach isotopes of shorter halflives are chosen allowing their halflives to be measured directly.

The two isotopes $^{221}$Fr and $^{224}$Ra (beta-decay daughter of $^{223}$Fr) have been chosen due to their suitable halflives, relative high yields and the clean beams conditions for this mass range of Fr isotopes from a W surface ion-source (for heavier masses RaF may become a problem).

<table>
<thead>
<tr>
<th>$t_{1/2}$</th>
<th>$^{221}$Fr</th>
<th>$^{224}$Ra</th>
<th>$^{224}$Fr</th>
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<tbody>
<tr>
<td>UCx SC yields</td>
<td>4.9 m</td>
<td>3.66 d</td>
<td>3.3 m</td>
</tr>
<tr>
<td>ThC SC yields</td>
<td>$2.8 \times 10^7/\mu$C</td>
<td>$1.4 \times 10^6/\mu$C</td>
<td>$4.7 \times 10^5/\mu$C</td>
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The two isotopes both have several decay daughters but none which poses problems neither as a contamination nor in the selection of the corresponding alpha-particle energy. In some cases the daughter decay, if sufficiently fast (e.g. $^{217}$At from the decay of $^{221}$Fr), can be used as a cross-check of the obtained halflife. Furthermore in the case of $^{221}$Fr the daughter decay of $^{213}$Bi (via alpha decay of $^{213}$Po) can be used to set the absolute time scale due to its very well known halflife.

Preliminary results on the decay halflife at room temperature of $^{221}$Fr performed at ISOLDE show a reduction of the halflife of 0.3 ± 0.2% in Au and of 0.42 ± 0.22% in W compared to Si. This is not yet conclusive, but indicates that our measurement should aim for a final precision better than $10^{-3}$. These preliminary results shown that the alpha decay of $^{221}$Fr is a very clean probe for halflife changes, see fig. 1.

The required count rate from the collected samples should be $10^2 - 10^3$ counts/sec at the beginning of data taking to maximize the collected statistics and limit deadtime. With a solid angle of 1% this corresponds to a $^{221}$Fr source of $5 \cdot 10^6 - 5 \cdot 10^7$ atoms, and a $^{224}$Ra source (produced from decay of $^{221}$Fr) of $5 \cdot 10^6 - 5 \cdot 10^8$ atoms.

The relative statistical accuracy that will be obtained for a single $^{221}$Fr sample will be a few times $10^{-3}$. We therefore need to repeat the measurement of each $^{221}$Fr sample 4-10 times to get the required $1 \cdot 10^{-3}$ accuracy. For $^{224}$Ra one measurement should be sufficient as long as the decay is followed for a sufficiently long time.

The alpha-particle energy could also be modified at the keV level by electron screening effects [17]. Measuring this with high precision would therefore be very interesting. This should easily be possible with Si surface-barrier detectors at room temperature where the energy resolution can be made smaller than 25 keV FWHM implying that energy shifts of a few keV would be detectable. The setup for the $^{221}$Fr decay, operating at room temperature, will consist of two Si detectors placed perpendicular on either side of a sample ladder with solid angles of approximately 1%. This setup will need to be on line e.g. at
the GLM line. For the $^{224}$Ra measurement collections should be made at the ISOLDE HV platform and measurements performed at an offline station with the same setup as described above.

It is important also to search for temperature dependences, similar to those observed for d+d reactions [5]. We plan to carry out a series of measurements at room temperature and with NICOLE operating at 4 K and 10 mK. Note that since we in all cases perform short collections and follow the decays for more than ten half-lives, the NICOLE and room temperature measurements can easily be performed in parallel. The total time needed is therefore determined by the measurement series at NICOLE.

For $^{221}$Fr we will need 1.5 shifts per material and temperature at NICOLE to perform 6 measurements to sufficient statistical accuracy. Firstly we would do the implantation in an insulator (Si, Diamond or Sapphire) at 4 K, here it might not be possible to go lower in temperature. Secondly we would change to the Au sample which takes 1 shift (Au is non-magnetic and will not give orientation, contrary to the experiments with Fr in Fe that were performed at ISOLDE in the past, e.g. [19]). We would perform two measurement series for Au at 4 K and 10 mK taking in total 3.5 shift, including 0.5 shift to cool down to 10 mK.

Whether we will measure the decay of $^{224}$Ra only at 4 K or at both 4 K and 10 mK will depend on the sample activity and on the outcome of the on-line $^{221}$Fr results. At minimum 3 samples for $^{224}$Ra will be needed: 1 Au at 10 mK and/or 4 K, 1 Au at 290 K and one insulator at 290 K.

For the half-life measurements in the NICOLE set-up Si p-I-n diode particle detectors with a sensitive thickness of 0.3 mm and a sensitive surface of 250 mm$^2$ will be used to detect the alpha particles. These will be operating with a solid angle of 1%, at a temperature of about 10 K inside the 4 K cooled part around the sample. The energy resolution under these conditions is typically about 30 keV FWHM at 6 MeV. Due to the limited cooling power at NICOLE at 10 mK of 1 microW one should be careful not to implant too strong sources. The required count rates of $10^3$ corresponds to 0.1 microW if all alpha particles (6.5 MeV) are stopped in the NICOLE fridge. It is therefore crucial to check the yields and impurities of the beam beforehand.
Summary and Beam-time request

We propose to measure with high accuracy the half-lives of several alpha decaying nuclei at different temperatures and in different materials.

For the production of the alpha-decaying nuclei we propose to use either a UC or ThC target with a W surface ionizer. The ThC target would be preferred since it has been shown at the SC to give higher yields of all isotopes of interest. Since it is not given that the SC yields are directly comparable to PS-booster yields we use the more conservative UCN yields from SC to calculate the beam-time request. The target should preferably be fitted with a heavy mass marker unless the stable U or Th comes out with high enough yields.

We also ask for 2 shifts at the beginning to optimize the purity of the source since neighboring masses have been seen in the preliminary data set [18], and to get yields of $^{224}$Fr so that the detailed timing of the measurements can be settled.

Summarizing we ask for:

- Optimization of beam purity 2 shifts.
- Changing sample and temperature in NICOLE 1.5 shifts.
- $^{221}$Fr beam on Si at 4 K 1.5 shifts.
- $^{221}$Fr beam on Cu at 4 K 1.5 shifts.
- $^{221}$Fr beam on Cu at 10 mK 1.5 shifts.
- $^{224}$Fr collections at HV platform 2 shifts.
- $^{226}$Ra collections at HV platform 2 shifts.

Furthermore we request the ISOLDE VME data acquisition and the CERN data storage system.

In total we ask for 12 radioactive shifts and 1 shift of beam tuning for the proposed experiment.

References