EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

Proposal to the ISOLDE and Neutron Time-of-Flight Committee

Probing two-alpha radioactivity with ^{224}Ra

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Abstract

The experiment aims at discovering the double alpha radioactivity, which has been recently predicted by members of the present collaboration. The ^{224}Ra candidate will be tested. The experimental signature would be the detection of two alpha-particles emitted back-to-back whose sum energy is 12.19 MeV. The branching ratio for this process is predicted to be around 10^{-9}, which implies special attention regarding the production rate and random correlations. We propose to implant ^{224}Ra nuclei in a thin foil surrounded by highly segmented Si detectors. Approximately 14 events are expected during a one-week beam time.

Requested shifts: 23 shifts (no split)

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Motivation

Detecting a new type of radioactivity is a high stake in the physics of the atomic nuclei. Recently, such a radioactive decay mode was predicted by members of the present collaboration [1]. It consists of the symmetric, back-to-back and simultaneous emission of two alpha particles by a heavy nucleus.

The emission of two alpha particles from a single nucleus has already been predicted since several years in a semi-microscopic approach. However, this mode was described as a ^{8}Be-like particle emission [2,3], yielding partial half-lives out of reach of current detection systems. For instance, a partial half-life of 10^{28} s was predicted for ^{224}Ra.

The present predictions rely on a more robust model, namely the covariant energy density functional (cEDF) approach, which has been successfully used in the last decade to describe both cluster phenomenology [4,5] and fission half-lives [6]. Recently, such a model was shown to properly reproduce half-lives for single-alpha decay (Table 1). This supports the validity of the present approach, which is based on the least-action path through potential energy (hyper)surfaces parametrized by the quadrupole, octupole and hexadecapole collective coordinates. It also provides a bridge between the descriptions of fission and alpha emission.

The same approach allowed the recent prediction of the simultaneous emission of two alpha particles, in opposite directions. The interesting point is that the corresponding predicted half-lives are within the reach of current detection setups, namely of the same order as cluster radioactivity half-lives. In the case of ^{224}Ra, the predicted two-alpha decay partial half-life is 10^{14} s.

Table 1: Comparison between the covariant-EDF predictions [1,7] and measurements [8,9] of lifetimes for single-alpha decay.

	^{104}Te	^{108}Xe	^{212}Po	^{224}Ra
T_{exp}	<18ns	58 μs	0.3 μs	3.6 d
T_{theo}	197ns	50 μs	0.6 μs	9.5 d

Guided by these predictions, experimental groups are attempting to discover the two-alpha radioactivity. Some of the authors of this proposal are involved in an experiment at GSI using a ^{228}Th source (the precursor of ^{224}Ra) and the FRS ion catcher. The present document is, to our knowledge, the first proposal for an in-beam study. Compared to the GSI measurement, it will benefit from a higher ^{224}Ra production rate and a high-performance setup.

Experimental approach

ISOLDE is an ideal installation to search for two-alpha emission as it is capable of delivering ^{224}Ra with a sufficient intensity (see e.g. the study of pear-shaped nuclei [10]). As mentioned above, the predicted branching ratio for the two-alpha decay is ~10^{-9}. Our goal is to observe the decay of at least 10^{10} ^{224}Ra nuclei which is perfectly feasible with the intensity delivered by ISOLDE. According to the ISOLDE yield database, ^{224}Ra can be produced with a yield of 6 10^{8} ions/μC using a UCx target. As we discuss below, in order to minimize random coincidences in our setup, we propose to run at reduced beam intensity, which will still provide a reasonable statistics.

We propose to implant the ^{224}Ra nuclei in a thin foil and detect their decay using highly segmented Si detectors belonging to the MUSETT and MUST II collaborations. The basic idea is to detect in coincidence two alpha particles emitted back-to-back. A box configuration of the setup is therefore proposed, as sketched in Figure 1. While the model predicts a back-to-back alpha emission, it does not provide any details concerning energy sharing. However, as the system is reflection-symmetric along its deformation path toward scission (see [1], Fig. 3), one can reasonably assume that the energy is shared almost equally between the two alpha particles. In this particular case, it is also possible to neglect the daughter recoiling energy, which results in the sum energy $E_{sum} = E_{2\alpha}(^{224}\text{Ra}) = Q_\alpha(^{224}\text{Ra}) + Q_\alpha(^{220}\text{Rn}) = 12.19$ MeV.

The signature of the two alpha decay is therefore a back-to-back detection of two alpha particles with a sum energy of 12.19 MeV.

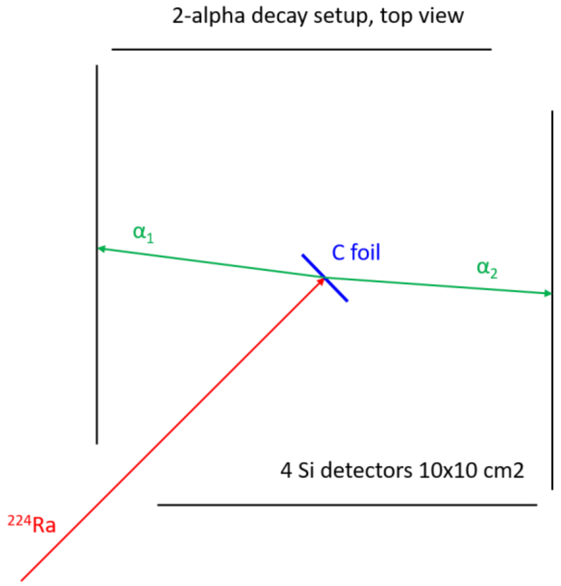

Figure 1: Concept of the two-alpha decay experiment.

Statistics and random correlations

Since the predicted branching ratio is very low, it is crucial to evaluate the random coincidence rate to assess the feasibility of the experiment. The decay chain of ^{224}Ra and its progeny is displayed in Figure 2.

The chain includes six decays ending with ^{208}Pb. The total activity is therefore 6 times the implantation rate in a steady-state regime (the secular equilibrium time is ~6 days and the maximum activity of the chain is reached after ~1.1 d). The implantation rate has to be a compromise between the final statistics, the detector counting rate capability and the random coincidence rate.

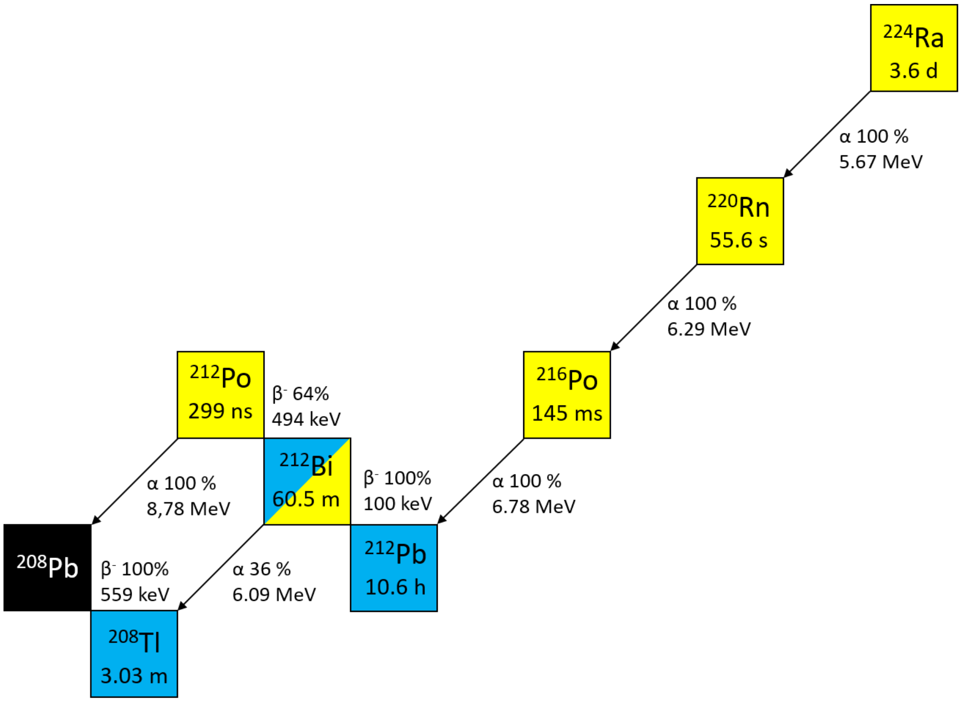

Figure 2: ^{224}Ra decay chain. For β emitters the average decay energy is provided.

Assuming a 5 10^4 pps ^{224}Ra implantation rate compatible with our setup performances (see below), about 30 two-alpha decays are expected after one-week beam time, out of which ~14 should be detected. There are two β decays in the chain. The thresholds of individual Si detector strips can be set high enough to cut off 75% of the beta particles such that in average 4.5 decays per implantation would contribute to the total counting rate. The random coincidence rate at which two opposite detectors would fire within a 100 ns coincidence window is estimated to be 350 Hz for the full array. If we require in addition that the two firing pixels are located back-to-back in a cone of 4 deg. half-aperture, this rate decreases to 0.43 Hz. We will use in addition a gate on the sum energy around $E_{2\alpha}(^{224}\text{Ra}) = 12.19$ MeV. The gain can be estimated from an experiment on the ^{224}Ra production and spectroscopy performed recently at IGISOL, University of Jyväskylä. Like in the present proposal, the ^{224}Ra nuclei were implanted in a thin foil placed between two Si pad detectors. From these data, we can estimate that random coincidences can be reduced by at least a factor 10^5 by gating on the sum energy, resulting in a random coincidence rate of 4.3 10^{-6} Hz for the full array.

We should also consider a possible pile-up in each pixel such that the measured energy using a 1µs peaking time is wrong, bringing the two-pixel summed energy to the expected range for the true two-alpha decay. We have to consider here the six radioactive decays following each ^{224}Ra implantation, leading to a maximum pile-up rate of ~3 10^{-5} Hz per pixel. Moreover, there are two β decays in the chain that correspond to a low energy deposition and consequently a smaller effect on the pile-up than alpha decays. If we consider that another alpha particle should be detected back-to-back within a maximum 100 ns time window, the pile-up coincidence rate is orders of magnitude lower than the random coincidence rate discussed in the previous paragraph.

Overall, a true-to-false double-alpha decay ratio of ~5 is estimated assuming a 5 10^4 ^{224}Ra implantation rate. If conditions prove to be clean enough (smaller coincidence time window, lower background using the sum energy window) we envisage increasing the beam intensity up to 2 10^5 pps. Rates are summarized in Table 2 for different experimental conditions.

Table 2 : Expected statistics and rates for different experimental conditions.

^{224}Ra beam intensity [pps]	5 10^4	10^5	2 10^5
Expected measured double-alpha decays, 7 days	14	27	54
Total counting rate [Hz]	10^5	2 10^5	4 10^5
Max. strip counting rate [Hz]	300	600	1200
Random correlation rate, 100 ns (30 ns) coincidence window [Hz]	340 (100)	1.3 10^3 (400)	5.5 10^3 (1.6 10^3)
True / False double-alpha decay ratio (time window, E_{sum} gate background reduction)	5 (100 ns, 10^5) 17 (30 ns, 10^5) 175 (30 ns, 10^6)	3 (100 ns, 10^5) 9 (30 ns, 10^5) 88 (30 ns, 10^6)	1 (100 ns, 10^5) 4 (30 ns, 10^5) 44 (30 ns, 10^6)

Experimental setup

We propose to use segmented Double-Sided Silicon Strip detectors from the MUST II [11] and MUSETT [12] arrays, arranged in a new configuration. The minor differences between these detectors and related electronics are not significant for the present proposal. Each Si detector has a size of 10x10 cm^2, is 128-fold segmented on each side and 300 µm thick.

As shown in Figure 1, the array will consist of four detectors arranged in an open box configuration, with the detectors positioned 60 mm from the source. The total efficiency for detecting an alpha particle (excluding inter-strip events) is approximately 45%. Since we are looking for two alpha particles emitted back-to-back, the efficiency is the same.

The ^{224}Ra nuclei with an energy of 30 keV will be implanted in a 200 nm C foil or alternatively 100 nm silicon nitride (Si3N4).

The array will be installed in a chamber provided by the collaboration.

The remotely controlled front-end electronics is based on ASICs installed in the vacuum chamber. The detectors and front-end electronics are cooled to maintain good performance and dissipate the electronics power. After shaping, the time and energy amplitudes are processed by VXI back-end electronics using “MUVI” and other cards for the clock distribution, trigger and read-out. The acquisition uses a Linux PC and the GANIL DAQ system. The cryothermostat is monitored by a Windows PC. All the electronics, power supplies, cooling system and DAQ can run standalone and can be fully remote controlled.

Note that the portability of MUST II and MUSETT electronics and DAQ has been demonstrated by campaigns at GANIL [13] and RIKEN, as well as measurements at the Saclay and Orsay detector labs.

During the run, the electronics will trigger on two-fold coincidences since our system cannot handle the ~100 kHz expected for 5 10^4 implantations per second. As explained above, the random coincidence rate for two opposite detectors responding within a 100 ns time window should be at the level of 350 Hz, which can be managed by the system with negligible dead-time. A fraction of single events will also be recorded.

The setup will be installed at the LA1 beam line. The beam intensity will be monitored by inserting periodically a fresh implantation foil.

Conclusion

The experiment aims at the first observation of double alpha radioactivity. We selected ^{224}Ra as a candidate with a predicted branching ratio of 10^{-9}. With a 5 10^4 ^{224}Ra implantation rate and an array of four highly segmented Si detectors, we expect to detect ~14 double-alpha decay candidates in a one-week beam time. If the conditions prove to be clean enough, beam intensity up to 2 10^5 pps will be used. Since ^{224}Ra has a half-life of 3.6 d, the measurement should continue at least 4 days after the beam period to accumulate further statistics.

Summary of requested shifts:

As a summary, 23 shifts of ^{224}Ra beam are requested with a maximum beam intensity up to 2 10^5 pps. This includes 2 shifts for tuning the beam. An offline measurement of at least four days after the beam period is also requested.

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Appendix

DESCRIPTION OF THE PROPOSED EXPERIMENT

The experimental setup comprises: *(name the fixed-ISOLDE installations, as well as flexible elements of the experiment)*

Part of the Choose an item.	Availability	Design and manufacturing
MUST II, MUSETT	☒ Existing	☐ To be used without any modification ☒ To be modified

HAZARDS GENERATED BY THE EXPERIMENT

Additional hazards:

Hazards	*MUST II / MUSETT setup*	*[Part 2 of the experiment/equipment]*	*[Part 3 of the experiment/equipment]*
Thermodynamic and fluidic			
Pressure	[pressure][Bar], [volume][l]		
Vacuum	< 10^{-6} mBar		
Temperature	253 K		
Heat transfer			
Thermal properties of materials			
Cryogenic fluid	Car cooling liquid, 0.8 Bar, 8 l		
Electrical and electromagnetic			
Electricity	120 V, 10 μA		
Static electricity			
Magnetic field	[magnetic field] [T]		
Batteries	☐		
Capacitors	☐		
Ionizing radiation			
Target material	^{12}C, Si3N4		
Beam particle type (e, p, ions, etc)	^{224}Ra		
Beam intensity	2 10^5 pps		
Beam energy	30 keV		
Cooling liquids	[liquid]		
Gases	[gas]		
Calibration sources:	☒		
• Open source	☐		
• Sealed source	☒ [ISO standard]		
• Isotope	^{239}Pu,^{241}Am, and ^{244}Cm		
• Activity	< 100 kBq		
Use of activated material:			
• Description	☐		
• Dose rate on contact and in 10 cm distance	[dose][mSV]		
• Isotope			

- Activity			
Non-ionizing radiation			
Laser			
UV light			
Microwaves (300MHz-30 GHz)			
Radiofrequency (1-300MHz)			
Chemical			
Toxic	[chemical agent], [quantity]		
Harmful	[chemical agent], [quantity]		
CMR (carcinogens, mutagens and substances toxic to reproduction)	[chemical agent], [quantity]		
Corrosive	[chemical agent], [quantity]		
Irritant	[chemical agent], [quantity]		
Flammable	[chemical agent], [quantity]		
Oxidizing	[chemical agent], [quantity]		
Explosiveness	[chemical agent], [quantity]		
Asphyxiant	[chemical agent], [quantity]		
Dangerous for the environment	[chemical agent], [quantity]		
Mechanical			
Physical impact or mechanical energy (moving parts)	[location]		
Mechanical properties (Sharp, rough, slippery)	[location]		
Vibration	[location]		
Vehicles and Means of Transport	[location]		
Noise			
Frequency	[frequency],[Hz]		
Intensity			
Physical			
Confined spaces	[location]		
High workplaces	[location]		
Access to high workplaces	[location]		
Obstructions in passageways	[location]		
Manual handling	[location]		
Poor ergonomics	[location]		

0.1 Hazard identification

3.2 Average electrical power requirements (excluding fixed ISOLDE-installation mentioned above): *(make a rough estimate of the total power consumption of the additional equipment used in the experiment)*

Total consumption is estimated at 10 kW.