EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

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

Properties of neutron-rich hafnium high-spin isomers

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Abstract

It is proposed to study highly excited multi-quasiparticle isomers in neutron-rich hafnium (Z = 72) isotopes. Long half-lives have already been measured for such isomers in the storage ring at GSI, ensuring their accessibility with ISOL production. The present proposal focuses on (i) an on-line experiment to measure isomer properties in ^{183}Hf and ^{184}Hf, and (ii) an off-line molecular breakup test using REXTRAP, to provide Hf^{+} beams for future laser spectroscopy and greater sensitivity for the future study of more neutron-rich isotopes.

Requested shifts: 9 on-line shifts (in 1 run) + 2 shifts of off-line test time (separately).

Introduction

Long-lived isomers in exotic nuclides give key structure information, which could be important for understanding heavy-element nucleosynthesis [1,2], and, more generally, the limits of nuclear stability [3]. Considerable progress has already been made using projectile fragmentation reactions, combined with the observation of γ-ray decays from microsecond isomers, see e.g. [1]. However, the γ-ray detection gives no experimental sensitivity to isomers with half-lives greater than 1 s, due to the need to time-correlate the arrival of each fragment with its subsequent γ-ray decay. The present work seeks to exploit spallation reactions combined with the ISOL method to study long-lived ($T_{1/2}$ > 1 s) isomers.

Following an accepted Letter of Intent [4] we now propose two ways forward: (i) to focus on isomers in ^{183}Hf and ^{184}Hf, which can be studied by γ-ray spectroscopy using established techniques; and (ii) to test the efficacy of using molecular breakup in REXTRAP, paving the way to the more neutron-rich hafnium isotopes and, at the same time, opening up the possibility of collinear laser spectroscopy. While the Letter of Intent [4] has not yet resulted in specific yield measurements of the more neutron-rich hafnium isotopes, it is through it that the planning of the present proposal has come about.

Motivation

The hafnium isotopes are well known [5] to contain highly excited, long-lived isomers, such as the 2.4-MeV, 31-year yrast trap in ^{178}Hf, with $K^{\pi} = 16^{+}$. This remarkable case has attracted considerable attention and controversy with regard to its potential as an energy-storage medium [6]. Nevertheless, the exceptional "yrastness" of that isomer is probably not unique. Nilsson [5,7] and Woods-Saxon-Strutinsky [8] model calculations indicate an even more favoured $K^{\pi} = 18^{+}$ isomer in ^{188}Hf. The present work addresses the experimental challenge to identify and characterise isomers in this region. To illustrate the energy favouring, calculated four-quasiparticle hafnium isomer energies, relative to rigid-rotor energies, are shown in Fig.1.

Notwithstanding the basic experimental challenge, there are additional physics issues involved. A leading consideration is that the ^{188}Hf neutron number of N = 116 is at the critical point of a prolate-oblate shape instability [8,9,10]. This is predicted [8] not to alter the basic isomer structure itself. However, the states to which the isomer decays could be of oblate rotation-aligned character [11], which may significantly influence the isomer half-life. Furthermore, high-K isomers and their decays could provide a unique opportunity to study the high-spin structure in this special region, where both the protons and the neutrons are in the upper portions of their respective shells (Z = 50 – 82, N = 82 – 126). This feature reinforces the stabilisation of well-deformed collective oblate rotation. Initial experimental evidence has recently been found in ^{180}Hf for this phenomenon [12], which is predicted to become an increasingly pronounced feature in the more neutron-rich isotopes [11].

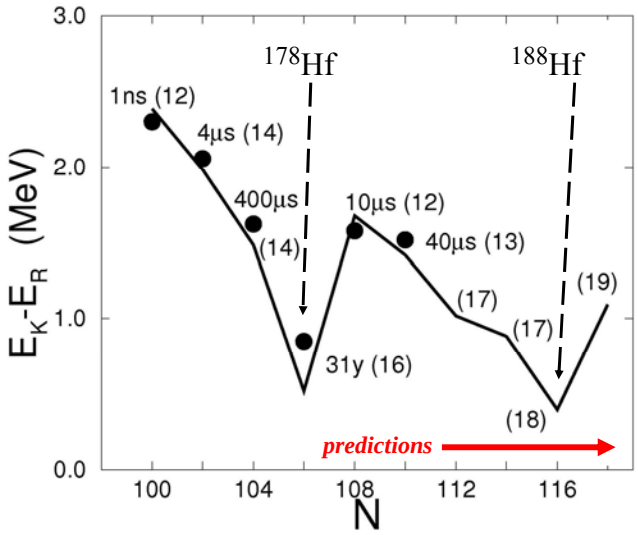

Fig.1. *Experimental (dots) and calculated (line) excitation energies relative to a rigid rotor of the same mass and spin, shown as a function of neutron number for four-quasiparticle isomers in even-even hafnium (Z = 72) isotopes. Spin values are given in parentheses, and experimental half-lives are shown. The figure is from ref. [7]. Note the strong empirical inverse correlation of excitation energy with half-life.*

A more general consideration is the need to understand isomer decay rates, since isomers can be longer lived than their respective ground states in exotic nuclei [3]. Indeed, due to the nature of experiments, it could be that various exotic nuclei are known only in isomeric states, rather than ground states. Further to these features, long-lived isomers can influence or reveal significant information about nucleosynthesis pathways [1,2]. We therefore consider that this neutron-rich, A ~ 190 region of long-lived isomers is a key testing ground for models of nuclear structure.

The specific measurements proposed here are motivated by new data [13] from the GSI Experimental Storage Ring (ESR), which demonstrate the existence of a long-lived (~12 min.) β-decaying isomer at 2.5 MeV in ^{184}Hf. This is the second isomer of ^{184}Hf. The first isomer (~50 s, 1.3 MeV) is known from earlier ISOL studies [14]. Examples of the ESR data are shown in Fig.2. While these data confirm some of the predictions [5,7], spectroscopic information is lacking. In addition, a ~10-s γ-decaying isomer was discovered [13] at 1.5 MeV in ^{183}Hf. A γ-decay event is illustrated in Fig.2 (right). These results give a clear objective for the initial phase of the present work: to measure the γ-ray emission properties from the recently discovered isomers in ^{183}Hf and ^{184}Hf.

In detail, the objectives of the proposed measurements are as follows:

(a) Establish the pathway of the β-delayed γ rays from 12-min. ^{184m2}Hf.

(b) Search for internal-decay γ rays from 12-min. ^{184m2}Hf.

(c) Establish the pathway of the internal-decay γ rays from 10-s ^{183m}Hf.

(d) Search for β-delayed γ rays from 10-s ^{183m}Hf.

The physics objectives of these particular measurements are to establish the isomer half-lives and decay pathways (level schemes) and hence determine the decay transition rates specific to each β or γ decay pathway. These will give measures of the K hindrance, and hence the degree of K mixing, which will test current interpretations that depend on the nuclear level density [15]. Whether or not the decreasing quadrupole deformation (with increasing neutron number) has a significant influence on the K mixing will be important for the general understanding of isomer stability, especially with the approach of the prolate-to-oblate shape/phase transition in the more neutron-rich hafnium isotopes.

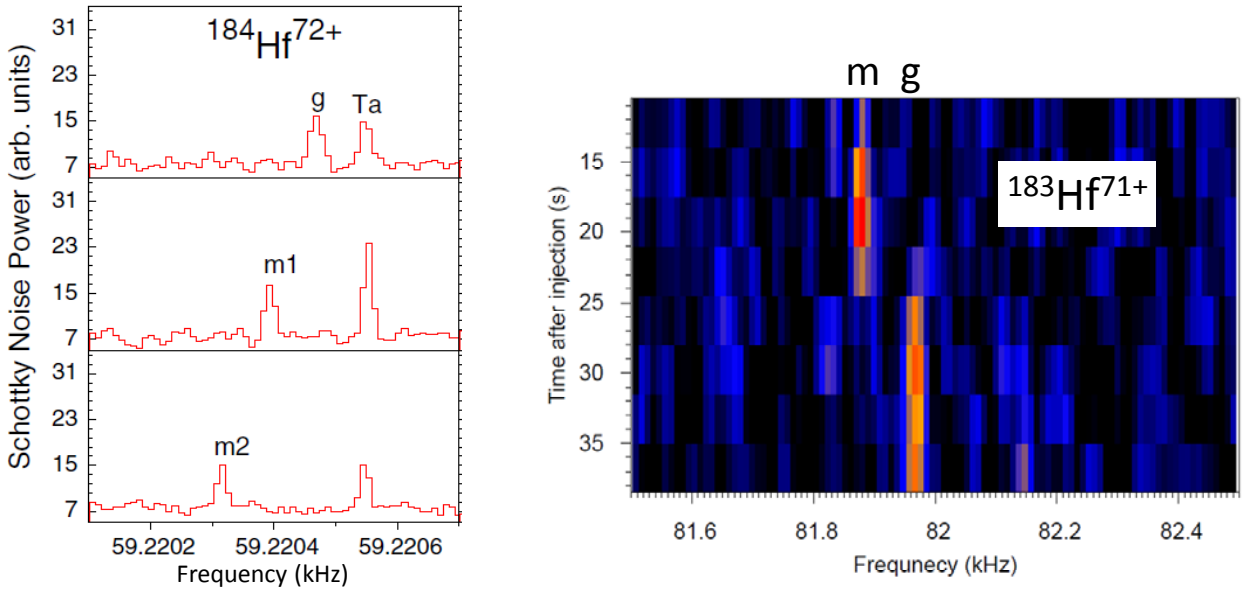

Fig.2. Left: *Examples of frequency spectra for A = 184 isobars and isomers, with different ions observed during three different injections in the ESR (taken from ref. [13]). Lower frequency corresponds to higher mass. The* ^{184}*Hf isomers (*m1 *and* m2*) and ground state (*g*) are identified as individual bare ions. The ground state of* $^{184}Ta^{72+}$ *is observed in each spectrum and provides as a convenient frequency reference.* ***Right:*** *Time resolved frequency spectra, showing initially a single q=71+ ion of* ^{183}*Hf in its isomeric state, which decays to the* ^{183}*Hf ground state about 1/2 way through the illustrated storage period.*

As already described in our associated Letter of Intent [4], these measurements will pave the way to (i) laser spectroscopy of Hf^{+} ions, enabling the deduction of isomer shifts, mean-square charge radii and electromagnetic moments, and (ii) access to more neutron-rich Hf isotopes and their accompanying isomers. The plan is to use REXTRAP to break up the fluoride molecular ions, and thus both purify the mass of interest, and at the same time create Hf^{+} ions that are suitable for laser spectroscopy. As a key step in this direction, we first propose to test off-line the breakup of hafnium (or tantalum) fluoride ions, as discussed in the next section. The two fluorides are expected to behave similarly in REXTRAP.

Experimental method

(i) On-line experiment

Radioactive hafnium beams, mainly as HfF_3^+, have been uniquely developed at ISOLDE [16]. Fluorination is required, using a CF_4 leak, to release the hafnium from the target. A Ta/W foil target is suitable for the neutron-rich cases highlighted in this proposal, i.e. ^{183}Hf and ^{184}Hf [16]. The addition of Ir foils, with extra neutrons and potentially improved release efficiency, has been used successfully for experiments at ISOLDE and is the preferred choice here. The isomeric decays of ^{183,184}Hf will be studied with a β/γ tape station on line LA1 (or LA2), by first implanting the isomer of interest for 2-3 half-lives, and then transporting the activity to a measuring station with a plastic scintillator for detection of β particles and conversion electrons, and two Ge detectors for γ rays. β-γ and β-γ-γ coincidences will substantially reduce background effects and enable detailed decay schemes to be constructed.

(ii) Off-line test

For more neutron-rich Hf isotopes, there is a complication from the complex molecules (oxy-fluorides) that are also formed. The difficulty is that the lighter Hf isotopes are much more strongly produced in the initial spallation reactions, and, in the form of complex molecules [16], they can contaminate the neutron-rich Hf beams. To solve this problem, we would like to inject mass-selected ions, e.g. $^{184}Hf^{19}F_3^+$ with A = 241, into REXTRAP for breakup of the molecular species. The method is similar to that described by Delahaye et al. [17] (though in that case they were studying the unbroken ions). After breakup, a time-of-flight mass analyser with an electrostatic kicker [17] can be used to select only the Hf^+ ions, rejecting all molecular ions. For example, $^{184}Hf^+$ (A = 184) will be clearly distinguishable from $^{184}Hf^{19}F^+$ (A = 203). It will thus be possible to produce exceptionally pure neutron-rich hafnium beams.

It is proposed that this aspect is first tested off-line, i.e. without a proton beam. Stable hafnium can be introduced into an ion source, together with a CF_4 leak, and the molecular breakup tested on HfF_3^+ ions. Alternatively, tantalum (as TaF_4^+ ions) could be used, as this will behave in a similar way and may offer more flexibility in scheduling. The successful testing of the molecular breakup will also open the way to collinear laser spectroscopy of Hf^+ ions.

Yield estimates for on-line experiment

Köster et al. [16] have compared experimentally obtained ISOLDE yields with different cross-section calculations for hafnium ground-state yields, though the situation for high-spin isomers is more complex. Two-proton removal from ^{186}W is a good way to populate ^{184}Hf [16], but the 2.5-MeV isomer will be produced with a significantly lower yield. The inclusion of Ir foils in the target is expected to give greater yield. (A ^{197}Au beam was used for

its discovery [13].) In the longer term, targets heavier than ^{186}W will certainly be needed, if heavier hafnium isotopes are to be produced. However, this is not a critical issue for the present proposal.

With $3x10^4$ ion/s of both ^{184g}Hf and ^{183g}Hf [16], in the form of HfF_3^+ ions, we estimate $3x10^2$ ion/s of ^{184m2}Hf and $3x10^3$ ion/s of ^{183m}Hf, i.e. with isomeric ratios in the range 1 – 10 %, which is consistent with other work [16]. The efficiency for transporting the activity to the measuring station will be about 25%, and the β and γ detection efficiencies will be close to 50% and 2% (for each Ge) respectively, giving a β-γ-γ rate of 1 per minute for ^{184m2}Hf multiplicity-two γ-ray cascades. Based on the need for 1000 counts in the strong transitions, 17 hours of continuous data taking are needed for ^{184m2}Hf. Although less time is, in principle, needed for the ^{183m}Hf measurement, in order to have a good probability of overall success we request 3 days of beam time – one for setting up, and one each for the two hafnium isomers.

Summary of requested shifts:

(i) On-line: 9 shifts (3 days) of ^{183}Hf and ^{184}Hf down line LA1 or LA2, from a Ta/W target, preferably including Ir foils.

(ii) Off-line: 2 shifts of HfF_3^+ (or TaF_4^+) through REXTRAP.

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Appendix

DESCRIPTION OF THE PROPOSED EXPERIMENT

The experimental setup comprises:

Part of the experiment	Availability	Design and manufacturing
Part 1: on-line experiment ISOLDE tape station 2 Ge detectors scintillator	X Existing	X To be used without any modification ☐ To be modified
	☐ New	☐ Standard equipment supplied by a manufacturer ☐ CERN/collaboration responsible for the design and/or manufacturing
Part 2: off-line test REXTRAP CENBG ToF beamline	X Existing	X To be used without any modification ☐ To be modified
	☐ New	☐ Standard equipment supplied by a manufacturer ☐ CERN/collaboration responsible for the design and/or manufacturing

HAZARDS GENERATED BY THE EXPERIMENT

(if using fixed installation) Hazards named in the document relevant for the fixed [COLLAPS, CRIS, ISOLTRAP, MINIBALL + only CD, MINIBALL + T-REX, NICOLE, SSP-GLM chamber, SSP-GHM chamber, or WITCH] installation.

Additional hazards:

Hazards	*ISOLDE tape station (on line)*	*REXTRAP and ToF (off line)*	*[Part 3 of the experiment/equipment]*
Thermodynamic and fluidic			
Pressure	standard ISOLDE vacuum	standard ISOLDE vacuum	
Vacuum			
Temperature	[temperature] [K]		
Heat transfer			
Thermal properties of materials			
Cryogenic fluid	LN2 for Ge detectors		
Electrical and electromagnetic			
Electricity	[voltage] [V], [current][A]		
Static electricity			
Magnetic field	[magnetic field] [T]		
Batteries	☐		
Capacitors	☐		
Ionizing radiation			
Target material	[material]		
Beam particle type	183Hf and 184Hf		
Beam intensity	30,000 /s		
Beam energy	30 keV		
Cooling liquids	[liquid]		
Gases	[gas]		
Calibration sources:			

• Open source	☐		
• Sealed source	X		
• Isotope	60Co, 152Eu, 133Ba		
• Activity	10 uCi		
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]		

3.1 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)*