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IN-BEAM AND DECAY STUDIES OF NEUTRON-DEFICIENT ISOTOPES OF HEAVY ELEMENTS*

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An intensive program to study the production, decay properties, and nuclear structure of isotopes of heavy elements is underway at the Department of Physics, University of Jyväskylä, Finland (JYFL). The main tools used in these studies are the gas-filled recoil separator RITU and various germanium detector arrays. Illustrative examples from both decay and in-beam studies will be presented.

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1. Introduction

The availability of large escape suppressed Ge detector arrays [1] has during the past few years made it possible to study the development of collective phenomena in nuclei far from stability. The established method of producing these nuclei is heavy ion induced fusion. In the region of heavy elements where fission of the compound system strongly dominates

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over particle evaporation, efficient filtering is needed to extract in-beam γ-rays originating from weak evaporation channels. This can be achieved by combining the Ge array with a recoil separator and by using the Recoil Decay Tagging (RDT) method [2]. Only those γ-rays will be accepted which are in coincidence with separated evaporation residues, identified through their characteristic decay in a position sensitive focal plane silicon detector. Most often, alpha decay is used for identification. An example of the power of the RDT method will be shown in Sect. 4 in connection with the discussion of the ^{184}Pb in-beam study.

Due to the selectivity of the process through which nuclear levels are populated by the emission of γ-rays following the production of evaporation residues, important non-yrast low lying levels are not always accessible to study using in-beam methods. A complementary method is then provided by radioactive α or β decay. An example is the observation of deformed $2p$–$2h$ intruder 0^+ states in the closed shell nuclei $^{192-198}$Pb from beta decay of Bi isotopes [3]. The determination of hindrance factors in alpha decay may also provide information on nuclear structure [4]. An example of such complementary studies will be discussed in Sect. 5 in connection with the alpha decay fine structure study of ^{192}Po.

The region of neutron-deficient nuclei around Pb with the closed $Z = 82$ proton shell is particularly interesting for the study of shape coexistence phenomena and the related intruder states. The closed proton shell favours spherical shape but around the midshell ($N = 104$) the interaction of $2p$–$2h$, $4p$–$4h$ and higher excitations across the shell with the large number of active valence neutrons leads to a delicate competition between spherical and deformed states at low spin and excitation energy [5,6].

In this paper, we will present results from in-beam and decay studies of neutron-deficient Hg, Pb, and Po nuclei performed at the Department of Physics, University of Jyväskylä, or JYFL. In addition, a preliminary report on results from an in-beam study of ^{254}No will be given. In all the experiments, the gas-filled heavy ion recoil separator RITU [7] was used to separate and identify the nuclei. Heavy ions were accelerated using the $K = 130$ MeV JYFL cyclotron.

2. The JUROSPHERE and SARI collaborations

During the year 1997, a series of in-beam γ-ray measurements were performed at JYFL using the JUROSPHERE array of escape-suppressed spectrometers. Some 3500 h of beam time were devoted to 25 experiments, most of them RDT-measurements. The Ge detectors were provided by the French/UK Loan Pool.

JUROSPHERE consisted of 15 Eurogam phase I detectors [8] and 10 TESSA detectors [9]. The Eurogam detectors have a relative efficiency of 70% (compared with a 76mm×76mm NaI(Tl) detector at 1.3 MeV) and were positioned at 134° and 158° relative to the beam direction. The TESSA detectors have a relative efficiency of 25% and were placed at 79° and 101°. The array had a total photopeak efficiency of $\sim$ 1.5% at 1.3 MeV. Typical beam intensities used were 5–15 pnA.

During the seven-month campaign, excited states were observed for the first time in several very neutron-deficient isotopes in the range $Z = 74$–92. Highlights from the project include structure studies of the following nuclides:

- ^{226}U from the reaction ^{208}Pb(^{22}Ne,4n) [10]
- ^{206}Ra from the reaction ^{170}Yb(^{40}Ar,4n) [11]
- ^{198}Rn from the reaction ^{166}Er(^{36}Ar,4n) [12]
- ^{184}Pb from the reaction ^{148}Sm(^{40}Ca,4n) [13]
- ^{168,170}Pt from the reaction ^{112}Sn(^{58,60}Ni,2n) [14]

Typical cross sections for the production of the most neutron-deficient nuclides were on the order of a few μb with the exception of ^{198}Rn for which the production cross section was about 200 nb.

In 1998, another set of detectors called SARI (Segmented Array at RITU) was assembled for a campaign during which some 1800 hours of beam time were devoted to 10 mostly RDT-experiments. The array consisted of three or four segmented clover detectors positioned around the target. The detectors were operated without Compton suppression shields but were protected by a thick layer of lead against radiation from the beam tube and the surroundings. The absolute efficiency of the array was 2–4% at 1.3 MeV depending on the number of detectors and on the target-to-detector distance. The set-up also involved Ge detectors at the RITU focal plane to detect γ-rays from long-lived isomers or from excited states populated through alpha decay. A close geometry system of four TESSA detectors with an efficiency of 0.8% was normally used. In two experiments, a large volume Ge clover on loan from GSI was used. Its efficiency was about 1.5%. In addition, a set-up consisting of three mini-orange type electron spectrometers around the target was used in some of the experiments.

Examples of RDT work performed using JUROSPHERE and SARI will be discussed in the following.

3. The shape of the nuclide ^{176}Hg along the yrast line

Mercury isotopes provide some of the best examples of shape coexistence. The known even–even Hg isotopes are weakly oblate ($\beta_2 \sim -0.15$) in their ground states, and their yrast structures display the corresponding oblate rotational bands down to ^{190}Hg. In lighter isotopes, the prolate intruder band with $\beta_2 \sim 0.25$ and with a larger moment of inertia becomes yrast at moderate spin. As expected, the prolate excited minimum reaches its lowest energy close to the mid-shell at $N = 102$ [15]. In very light even–even Hg isotopes, according to recent Nilsson–Strutinsky calculations [6], the ground state is expected to be spherical while the prolate structure might disappear or give way to a superdeformed structure with $\beta_2 \sim 0.5$.

The increase in the excitation energy of the prolate band, as one proceeds towards the proton drip line, was verified for ^{178}Hg in a recent RDT experiment [16] using the JUROSPHERE array. In the same work, three γ-ray transitions were tentatively assigned to an E2 cascade de-exciting the lowest 2^+, 4^+, and 6^+ states in ^{176}Hg. The question of the disappearance of the prolate minimum remained open, however. In the present work [17], we have performed an extended study of the level structure of ^{176}Hg, produced in the reaction ^{144}Sm(^{36}Ar,4n)^{176}Hg at a bombarding energy of 190 MeV. The estimated cross section was about 7 μb, and altogether 90000 ^{176}Hg α decay events with full α particle energy were observed.

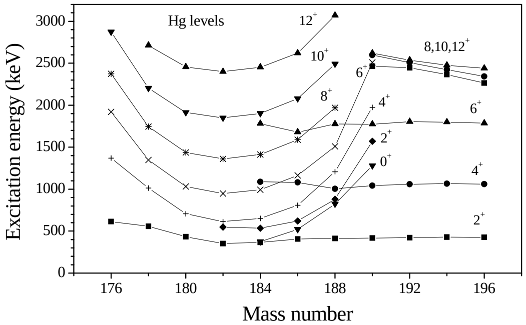

Fig. 1. Energy level systematics of neutron-deficient even-mass isotopes of Hg.

The tentative level scheme of ^{176}Hg, based on results from the present work, including RDT γ–γ coincidence data, confirms the assignments of Carpenter *et al.* [16] for the lowest three levels. The energy level systematics of a range of Hg isotopes is shown in Fig. 1. The rise of the lowest 2^+ and 4^+ level energies in ^{176}Hg is an indication of a transition towards a spherical ground state in accordance with predictions [6]. The compression of the energy differences between the positive parity yrast levels above the 6^+ and 8^+ state is interpreted to be the result of the crossing of the prolate band.

The two-band mixing model [18] was used to extract the undisturbed energy difference between the prolate and oblate band heads. Variable moment of inertia (VMI) parameters and the prolate-oblate interaction strength (~ 100 keV) which reproduced the ^{178}Hg level scheme [16] were used. A value of roughly 1300 keV was deduced for the energy difference. This is approximately 600 keV higher than in ^{178}Hg, revealing a rapid increase of the excitation energy of the prolate structure with decreasing neutron number.

4. First observation of excited states in ^{184}Pb

The occurrence of different shapes in one nucleus at low excitation energy and angular momentum is a well-known feature of neutron-deficient nuclides around $Z = 82$ [19]. Low-lying 0_2^+ states associated with weakly-deformed oblate structures have been observed to coexist with spherical ground states in several Pb isotopes with $N \geq 106$ [19,20]. These states are usually described as resulting from $2p$–$2h$ excitations across the shell gap. In ^{186,188}Pb, low-lying deformed rotational structures have been observed at $I > 2\hbar$ by Heese *et al.* [21] and Baxter *et al.* [22]. The bands, which are similar to those in the corresponding isotones ^{184,186}Hg, are associated with predicted [6] prolate-deformed minima. In the work described here [13], excited states have been observed for the first time in ^{184}Pb.

Excited states in ^{184}Pb were populated in the reaction ^{148}Sm(^{40}Ca, 4n)^{184}Pb at a bombarding energy of 195 MeV. Prompt γ-rays from the target were observed using the JUROSHERE array. Approximately 9000 full energy alpha particles from the 0.6 s ground state in ^{184}Pb were collected in the RDT-experiment.

The spectrum in Fig. 2(a) shows prompt γ-rays in coincidence with all recoils detected at the RITU focal plane. It is dominated by γ-rays from Hg isotopes produced in αxn evaporation channels and in fusion reactions between the contaminant ^{40}Ar beam (relative intensity approximately 5%) and the ^{148}Sm target. The power of the RDT method is seen by comparing the spectrum in Fig. 2(a) with that in Fig. 2(b) which shows those γ-rays identified as belonging to ^{184}Pb. A maximum time difference of 1.5 s *i.e.* approximately three ^{184}Pb half-lives was allowed between an evaporation residue and an 6.63 keV α particle.

The intensity of the peaks in the spectrum displayed in Fig. 2(b) does not allow a γ–γ coincidence study of ^{184}Pb. Assignment of γ-ray peaks to transitions within a rotational band is based on systematics and on relative intensities of the transitions. The resulting excitation energies are shown for the isotopes ^{184,186,188}Pb in Fig. 3. By using variable moment of inertia (VMI) fits it was possible to extract the prolate band head energies for the three isotopes ^{184,186,188}Pb. The following results were obtained: 610 keV

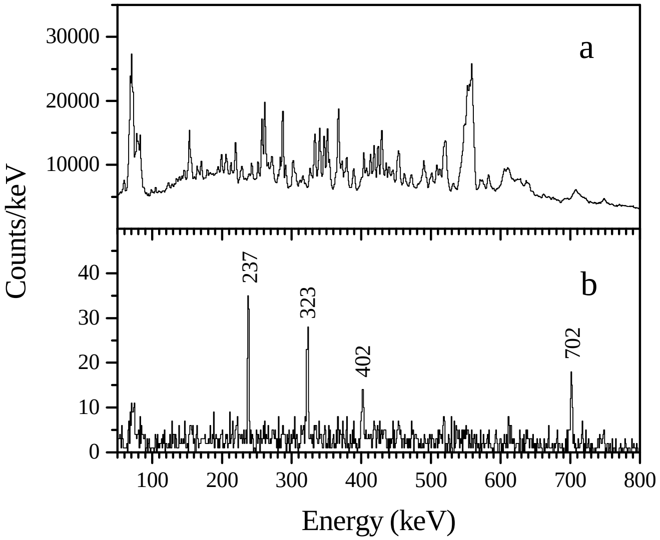

Fig. 2. (a) — Singles γ-ray energy spectrum from the reaction ^{40}Ca + ^{148}Sm. Gamma-rays in coincidence with all fusion evaporation residues detected at the separator focal plane have been accepted. (b) — The recoil-decay-tagged γ-ray spectrum of ^{184}Pb.

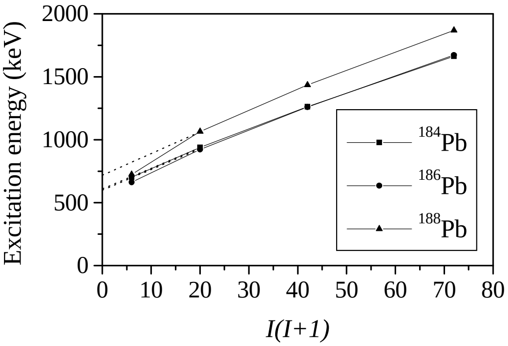

Fig. 3. Excitation energy as a function of spin I for ^{184,186,188}Pb. The dotted lines are extrapolations to zero spin (see text).

(^{184}Pb); 600 keV (^{186}Pb); 710 keV (^{188}Pb). The effect of mixing of states on the band head energies needs to be considered carefully (see Refs [13] and [23]). The result is that the prolate band head energies for ^{184}Pb and ^{186}Pb are nearly identical and considerably lower than that for ^{188}Pb. This indicates that the excitation energy of the prolate-deformed configuration has a minimum close to $N = 103$ as expected.

5. Fine structure in the α decay of ^{192}Po

As discussed in the preceding Section, prolate structures in the neutron-deficient ^{184}Pb, ^{186}Pb and ^{188}Pb nuclei were discovered recently in in-beam experiments [13,21,22]. It was concluded [21] that the structure of the observed bands is different from the $2p$–$2h$ proton intruder states in $^{190-208}$Pb mentioned above. Due to decay out of the bands, the 0^+ band heads were not observed in these experiments.

In another recent work [20], on fine structure in the α decay of ^{192}Po, an excited 0^+ state was observed in ^{188}Pb. It was concluded that this was the oblately deformed $2p$–$2h$ proton intruder state observed in heavier Pb isotopes.

In the work performed at JYFL [23], the standard position-sensitive focal plane detector [7] was combined with an auxiliary detector system for observing alpha particles and conversion electrons escaping from the stop detector at backward angles. The reaction used to study fine structure in the alpha decay of ^{192}Po was ^{36}Ar + ^{160}Dy. The bombarding energy was varied between 172 and 184 MeV and the target thickness was 500 μg/cm^2.

When alpha decay takes place from the ground state of ^{192}Po to low-lying excited 0^+ states in ^{188}Pb, the excitation energy in the daughter nucleus is expected to be removed primarily through conversion electrons from E0 transitions. Thus, by demanding a coincidence between alpha decays in the stop detector and conversion electrons in the auxiliary detector, it was possible to observe alpha decay fine structure to two low-lying 0^+ states in ^{188}Pb. One of these, at an excitation energy of (568±4) keV, was identified as the oblate band head thus confirming the result of Ref. [20]. The other state has an excitation energy of (767±12) keV and was identified as the prolate 0^+ band head.

It is of interest to deduce the mixing amplitude of the oblate $4p$–$2h$ state in the ground state of ^{192}Po. This can be achieved through experimental alpha decay hindrance factors and mixing amplitudes for the excited 0^+ states in ^{188}Pb [4]. The hindrance factors as determined relative to alpha decay to the ground state of ^{188}Pb were deduced to be 0.67±0.10 (decay to the oblate 0^+ state) and 0.22±0.08 (decay to the prolate 0^+ state). The mixing amplitudes were determined following the procedure of Dracoulis [24] by fitting to the experimental levels from this work and from Ref. [21]. Then, by representing the ground state of ^{192}Po as a combination of $2p$–$0h$ and $4p$–$2h$ states, it was possible to determine the admixture of $\sim 63\%$ for the intruder $4p$–$2h$ configuration in the ground state of ^{192}Po. The unmixed prolate 0^+ level energy of ~ 730 keV in ^{188}Pb agrees well with the value of ~ 710 keV estimated from the energies of higher members of the band [13,21] (see Section 4).

6. Observation of excited states in ^{254}No

Efforts aimed at the synthesis of new elements have, following a steady progress in detection techniques, lead to production of elements up to proton number 112 [25]. Methods such as He-jet techniques in combination with bombardment of actinide targets with relatively light ions lead to the synthesis of elements with $Z = 102$–106 [26]. The elements with $Z = 107$–112 were produced using the method of cold fusion [27] where Pb or Bi targets were bombarded with ions of Cr–Zn so that the 1n neutron evaporation channel was observed. To separate the new nuclei from unwanted particles, the fast and efficient method of in-flight separation with the on-line velocity filter SHIP [28] at GSI, Darmstadt, was used.

The ultimate goal of the study of the heaviest elements is not only to find the limit of existence of elements but also to search for the long-predicted island of spherical super heavy nuclei [26]. The main result from recent progress in this field is the discovery that the shell stabilized island is not separated from the peninsula of known nuclei by the sea of fission as originally expected. Rather, a region of enhanced stability against fission has been found. The reason behind this enhancement is microscopic stabilization, and the main decay mode for isotopes of the heaviest elements is alpha decay.

The rather neutron-deficient isotopes of the heaviest elements are expected, on the basis of theoretical calculations, to be deformed. The shell stabilization of the trans-actinides has been found to originate from a negative hexadecapole (β_4) deformation in the ground state [29]. As expected, the most important component in the deformation is the quadrupole deformation which has a relatively constant value of $\beta_2 \sim 0.24$ in a large region on the chart of nuclei centered around ^{256}No [29]. The relative importance of higher order terms depends on the nuclide.

Two of the most important goals of further studies in the region of the heaviest elements are experimental establishment of their deformation and deeper insight into their production mechanism in heavy-ion-induced fusion reactions. Measurement of the level energies of the ground state band of an even–even nucleus would provide valuable information on the deformation and might also give the first hints concerning the limitation on angular momentum values leading to the formation of evaporation residues.

The excited states of even–even nuclei in the region of quadrupole deformation around ^{254}No are characterized by closely spaced rotational bands where the first 2^+ energy is around 50 keV. This excitation energy has been measured for ^{248,254,256}Fm on the basis of α or β decay studies [30]. No data were known for No isotopes previously but there is an estimate of (51 ± 35) keV for the 2_1^+ energy of ^{256}Rf ($Z = 104$) from alpha decay of ^{260}Sg [31]. On theoretical grounds, no sudden changes in structure are expected for ^{254}No. There is a deformed neutron subshell closure ($N = 152$) at ^{254}No but its effect on energies of excited states is expected to be small. One can with rather good confidence estimate that the first excited levels of the ground state rotational band in ^{254}No are at about 50 keV (2^+), 150 keV (4^+), and 300 keV (6^+). The corresponding conversion coefficients for E2 transitions are ~ 1000, ~ 25, and ~ 6 for $2^+ \to 0^+$, $4^+ \to 2^+$, and $6^+ \to 4^+$ transitions, respectively. It is thus expected that the most likely gamma-rays to be observed in an in-beam experiment, taking into account the estimated fusion rate, are the $6^+ \to 4^+$ and $8^+ \to 6^+$ transitions.

Experiments to study excited states of ^{254}No have recently been performed in two laboratories. In the Argonne National Laboratory, the Gammasphere Ge detector array [32] coupled to the Fragment Mass Analyzer [33] was used in a successful experiment in July, 1998 [34]. In August, 1998, the SARI array together with RITU was employed in a three-week experiment. Results from this experiment will be given in the following. In both experiments, the reaction ^{48}Ca + ^{208}Pb was used. At JYFL, the excitation function was measured. The maximum cross section was determined to be about 2.1 μb at a bombarding energy of 216 MeV, corresponding to about 21 MeV excitation of the compound nucleus. This compares well with results from measurements at SHIP but is somewhat lower than values from experiments performed using methods of nuclear chemistry [35]. The beam intensity was typically 10 pnA, and the target thicknesses used varied between about 250 and 700 μg/cm^2. The counting rate of ^{254}No α particles with full energy was typically 20–40/h, and altogether about 12000 full energy alpha particles were observed in the experiment.

The recoil-gated and RDT gamma-ray spectra were somewhat affected by the Compton background resulting from the lack of Compton-suppression shields. Nevertheless, several γ-ray peaks assigned to ^{254}No were observed, in general agreement with data from the Argonne experiment. In addition, a new γ-peak with an energy of ~ 414 keV, tentatively assigned to the $16^+ \to 14^+$ transition, was observed. The analysis of the data is in progress.

7. Discussion

We have presented results from both decay and in-beam studies performed mainly in the region of very neutron-deficient isotopes ranging from Hg to Po. The combination of γ-ray detectors with a recoil separator has been fruitful in this work because of rather favourable experimental conditions. Reaction cross sections, typically on the order of a few μb, are still sufficient for RDT studies and the half-lives, well below one second, are well suited to the correlation technique. The continuation of these studies to even more neutron-deficient isotopes will be difficult due to the rapidly decreasing cross sections.

In the case of Hg and Pb isotopes, new data have been collected on rotational structures. The systematics for ^{184,186,188}Pb [13] concerning the excitation energy of the prolate band head is well in line with the result for ^{188}Pb based on our alpha decay fine structure study [23].

An interesting example of the kind of complementary information one can deduce from decay experiments is our value for the admixture of the oblate $4p$–$2h$ component in the ground state of ^{192}Po as deduced on the basis of alpha decay hindrance factors, $\sim 63\%$ [20,23]. On the basis of Nilsson–Strutinsky type of calculations it was suggested by May *et al.* [5] that in ^{192}Po an oblate deformed minimum becomes the ground state. The result is also compatible with that from our RDT in-beam work on levels of ^{192}Po [36].

The pair of experiments on the structure of ^{254}No [34,37] have accomplished a major step towards a deeper understanding of the physics of very heavy nuclei. The progress was made possible by the exceptionally high cross section in the reaction employed, ^{48}Ca + ^{248}Cm. There is now, for the first time, direct experimental evidence on both the shape of a transfermium nuclide and the amount of angular momentum such a nucleus can sustain. Because the internal conversion coefficients of the low-lying ground state band transitions are high, improvement on the data can be expected once recoil decay tagged conversion electron measurements, presently at the planning stage at JYFL, can be realised. It would also be of great importance to gain knowledge on single particle energies from studies of the odd nuclides in this region. These require considerable improvements in the overall efficiency of the experiments, however, since the cross sections for producing the nuclides ^{253,255}No and ^{255}Lr are about one order of magnitude lower [35] than that for the reaction leading to ^{254}No used at JYFL and Argonne. Further reductions in γ-ray transition intensities may result from fractionated decay paths in the odd-mass nuclei.

RDT experiments will continue to be a major part of the work at JYFL. Experimental developments include a detector system that allows the use of beta decay for tagging of fusion products. Due to wide interest in nuclides produced in symmetric reactions, design of an improved separator with better background conditions but high transmission will be undertaken.

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