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Fine Structure in the alpha decays of ^{226}U and ^{230}Pu

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Abstract. The nuclei ^{226}U and ^{230}Pu have been populated via reactions involving ^{208}Pb targets bombarded by ^{22}Ne and ^{26}Mg projectiles. Fusion-evaporation residues were separated in-flight using a gas-filled recoil separator. A position-sensitive Si-strip detector was employed at the focal plane in order to identify correlated α-decay chains. Two fine structure α-decay lines have been observed. The first, with an energy of 7385(5) keV, is assigned as the α decay from ^{226}U to the first excited 2^+ state of ^{222}Th. The second line, observed for the first time in this work, has an energy of 6961(30) keV and is assigned as the α decay from ^{230}Pu to the first excited 2^+ state of ^{226}U. The excitation energy of the first excited 2^+ state in ^{226}U was determined to be 96(25) keV, giving an estimate of the deformation parameter $\beta_2 \simeq 0.20$.

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1 Introduction

Recently, the identification of excited states in ^{226}U was reported for the first time [1]. The γ-ray transitions were unambiguously assigned to ^{226}U through the recoil-decay tagging (RDT) technique [2,3]. Due to large internal conversion, and the low energy expected for the 2^+ to 0^+ transition in ^{226}U, the excitation energy of the 2^+ state could not be confidently determined from the γ-ray data obtained, though a candidate was observed with an energy of 80.5(5) keV [1]. In this paper, the results of an experiment to populate the aforementioned 2^+ state through the α decay of ^{230}Pu are presented. This measurement allows the absolute excitation energies of the excited states in ^{226}U to be determined, and provides an estimate of the β_2 deformation parameter. Also presented are improved measurements of the α-particle energies and branching ratios of the decay of ^{226}U into ^{222}Th. These fine structure measurements provide information which can be used to obtain improved values for quantities such as the reduced α-decay width and the hindrance factors for decays to excited states.

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2 Experiments

Two experiments have been performed, both employing the RITU [4] gas-filled recoil separator. Beams of ^{22}Ne and ^{26}Mg, from the K=130 MeV cyclotron situated at the Department of Physics of the University of Jyväskylä, Finland, bombarded self-supporting ^{208}Pb targets. Fusion-evaporation residues were magnetically separated in flight from unwanted products such as fission fragments and primary beam, and implanted into a Si-strip detector positioned at the focal plane of the RITU device. The detector is divided in the horizontal direction into 16 strips, sometimes coupled together to form 8 strips. Position sensitivity in the vertical direction is achieved through charge division. This position sensitivity in both the horizontal and vertical directions allows identification of correlated α-decay chains, using the method described in [5]. In both experiments, the resolution of the Si-strip detector was better than 30 keV FWHM for α decays with an energy of approximately 7.5 MeV. The helium gas used in RITU, typically at pressures of 0.5 to 1.5 mbar, was separated from the beam-line vacuum by a thin carbon(0.1 mg/cm^2, first experiment) or nickel(0.45 mg/cm^2, second experiment) foil.

In the first experiment, the ^{208}Pb(^{22}Ne,4n)^{226}U reaction was used (maximum cross-section $\simeq$7 μb [6]), with a total irradiation time of around 320 hours. The ^{22}Ne beam bombarded a ^{208}Pb target of thickness of 250 μg/cm^2 with an average intensity of approximately 16 particle nA. The centre of target beam energy was 111 MeV, taking into account the small energy loss through the thin carbon gas containment window. The transmission efficiency of RITU was estimated to be approximately 10% for the recoiling U nuclei. The main goal of this experiment was to identify excited states in ^{226}U through the RDT technique, as discussed in Sect. 1; results can be found in [1]. In addition to the focal plane Si-strip detector, a single TESSA-type [7] Ge detector was mounted behind the focal plane of RITU, in order to observe γ-rays emitted from states populated by α decay. Detected γ rays were only read into the data stream when occurring in coincidence with events in the Si-strip detector.

The second experiment, to populate the first excited 2^+ state of ^{226}U, employed the ^{208}Pb(^{26}Mg,4n)^{230}Pu reaction. In this case, the transmission efficiency of RITU was estimated to be approximately 15% for the recoiling Pu nuclei. A natural Mg compound was placed in the JYFL ECR ion source to enable extraction of a ^{26}Mg beam using the MIVOC technique [8]. The ^{26}Mg beam was then accelerated to 140 MeV by the K=130 MeV cyclotron. The energy loss through the Ni gas containment window was 2.4 MeV. A further single 0.9 mg/cm^2 Ni foil was then used to degrade the incident beam energy to around 132 MeV (centre of target), where fusion-evaporation code HIVAP [9] calculations predict the maximum cross-section for the reaction given above. The reaction cross-section at this energy, assuming a RITU transmission efficiency of 15%, was estimated from our data to be less than 50 nb. In this experiment the beam was macro pulsed with beam phases of 5 ms on/5 ms off and 8 ms on/2 ms off in order to reduce the contribution from background events. The average beam current after pulsing was approximately 30 particle nA. A rotating ^{208}Pb target was used with foil thicknesses varying between 500 and 800 μg/cm^2. The total irradiation time was approximately 70 hours. Since the states populated by α decay were expected to be of low energy, and the γ transitions would be strongly converted, no Ge detector was placed behind the focal plane of RITU.

3 Results

3.1 Alpha Decay of ^{226}U

Decay events associated with ^{226}U were selected by searching for position and time correlated triple α-α-α chains. The energy of the first event in the chain was allowed to vary freely, with the condition that it must be followed by two subsequent events in the same strip, with energies of (7942 to 8022) keV and (17.1 to 17.5) MeV, within a vertical position window of approximately $\pm$ 1 mm. The first energy range corresponds to the α decay of ^{222}Th, the second to the full energy sum peak of the fast decays of ^{218}Ra and ^{214}Rn. The maximum time difference between events of type α_1-$\alpha_{7.982}$ was 12 ms, and between events of type $\alpha_{7.982}$-α_{sum} was 0.15 ms. The spectrum of events extracted using these search criteria is shown in Fig. 1(a).

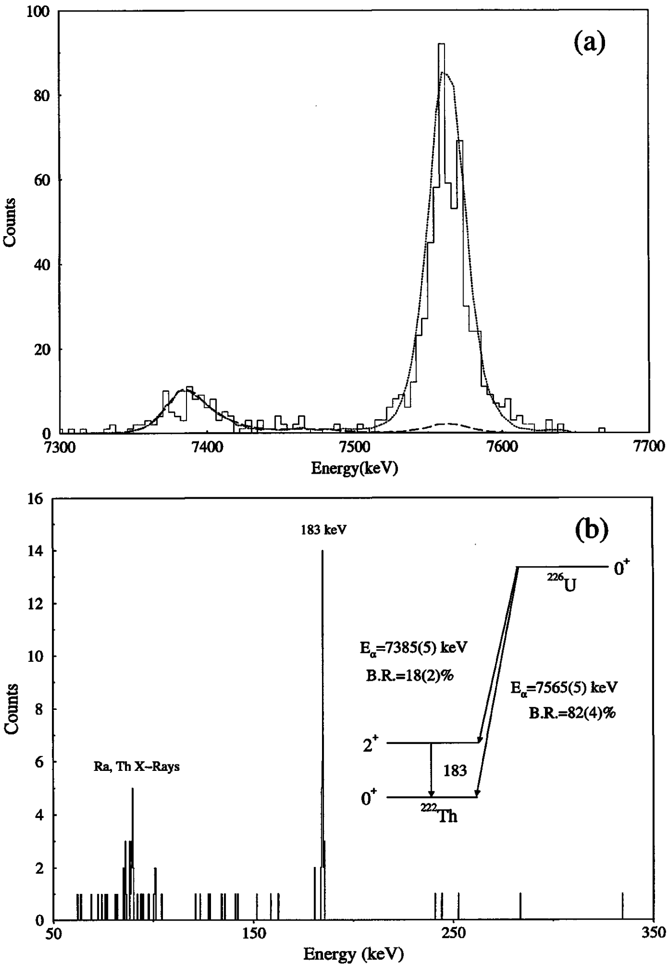

Fig. 1. (a)Events correlated with the α decay of ^{222}Th and full energy α-particle sum peak of ^{218}Ra + ^{214}Rn. The dotted and dashed lines are the results of a Monte Carlo simulation, see text for details (b) Gamma-ray events in coincidence with events of energy (7340 to 7430) keV in the focal plane Si-strip detector. The branching ratios and energies shown are those corrected for the summing effect

It can be seen that there are two distinct α groups. The more intense line has an energy of 7565(5) keV, consistent with the previously assigned ^{226}U ground state (g.s.) to ^{222}Th g.s. α decay [10]. The other α group has a measured energy of 7390(5) keV. This value is different to that measured by Andreyev et al (7420(20) keV), though the measured intensity ratios are similar (16(2)% (this work) 15(5)% (Andreyev et al [10])). Two α-decay lines with energies of 7566(20) and 7394(20) keV, with branching ratios of approximately 72 and 28%, respectively, were also reported by Enqvist et al. [11].

The measured time differences between these two α groups and events corresponding to the 7982 keV α decay of ^{222}Th, yield half-lives of 2.2(3) and 2.1(1) ms for the decays following the lines at 7390 and 7565 keV, respectively. These values are consistent with the half-life for the α decay of ^{222}Th quoted in [10] of 2.6(4) ms. The difference in energy of these two α-decay lines (175 keV) is similar to the known 2^+ to 0^+ transition energy in ^{222}Th (183 keV). The small difference in energy is due to the effect of conversion electrons partially summing their energy with the α decay energy. The recoil energy of the daughter nucleus also contributes to this shift. It should be noted that the energy shift due to complete summing of K conversion electrons is approximately 70 keV. Indeed, there is evidence in Fig. 1(a) of a peak corresponding to this complete summing at an energy of approximately 7458 keV. In order to quantify the summing effect, a Monte Carlo simulation has been employed. The simulation calculates the total energy deposited by a particular event, from knowledge of the following: the g.s. to g.s. α-particle energy, the excitation energy of the excited state, the relative branching ratios to the ground and excited states, the detector resolution and the implantation depth. The relevant internal conversion coefficients are taken from the tabulated values of Rösel et al [12]. The code treats the multiple scattering of electrons in the Si in the same manner as that described in [13]; emission of X-rays following internal conversion is also taken into account. Using a transition energy of 183 keV, the Monte Carlo code reproduced the experimentally measured α-particle energies to within 0.5 keV. The spectrum obtained from the Monte Carlo simulation is shown as the dotted line in the inset to Fig. 1(a). Thus, using the known ^{222}Th 2^+ to 0^+ transition energy of 183 keV, and taking into account the small difference in recoil energy terms for the α decays to the ground and excited states, the α-particle energy for the transition ^{226}U (g.s.) to ^{222}Th (2^+) is 7385(5) keV. This assignment is confirmed by the observation of a strong γ-ray peak of energy 183 keV in coincidence with Si-strip detector events of energy (7340 to 7430) keV, see Fig. 1(b). The observation of Ra X-rays in this figure is due to the tail of the ^{223}Th α-decay peak entering the coincidence gate. The dominant α decay line from ^{223}Th populates an excited $5/2^+$ state in ^{219}Ra, which then decays via a strongly converted 140 keV $M1$ transition to the $7/2^+$ ground state.

Also shown in Fig. 1(a) (dashed line) is a further output spectrum from the Monte Carlo simulation. This spectrum corresponds to those events which originate from population of the 2^+ state in ^{222}Th via the α decay of ^{226}U. It can be seen that the summing effect shifts a fraction of the events below the g.s. to g.s. α-decay peak. The measured intensity ratios quoted earlier must therefore be corrected in order to obtain the branching ratios. In this case, the fraction of decays to the 2^+ state which are shifted into the g.s. to g.s. peak is 13%. Thus the deduced branching ratios are 82(4)% and 18(2)% for the decays to the ground and 2^+ states, respectively.

Figure 2(a) shows a plot of $Q_\alpha^{-1/2}$ (where Q_α is the α decay Q-value) against the logarithm of the partial half-life for several U and Pu isotopes, yielding the well-known Geiger-Nuttall curves. In every case, the error bars are smaller than the symbol used. The Pu isotopes are dis-

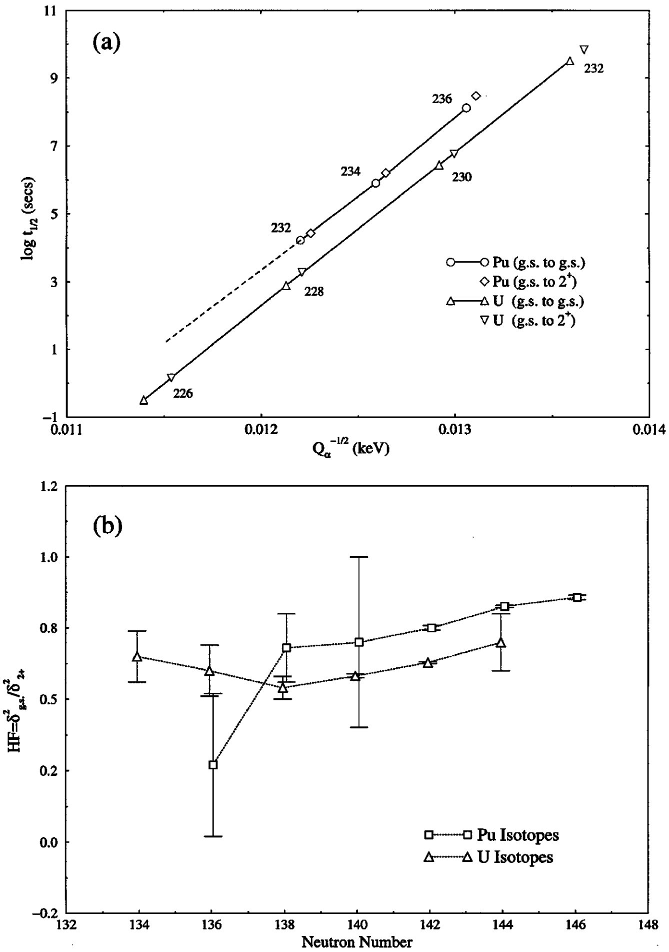

Fig. 2. (a) Plots of $Q_\alpha^{-1/2}$ against the logarithm of the partial half-life for several even-even U and Pu isotopes. The dashed line is an extrapolation used to determine the partial half-life for the g.s. to g.s. decay of ^{230}Pu.(b) Hindrance factors for isotopes of U and Pu calculated using the formulism of Rasmussen [15]

cussed in the following section. Data for the heavier U isotopes were taken from [14]. It can be seen that the data for ^{226}U deduced from this work ($Q_\alpha^{-1/2} \simeq 0.0115$ and $T_{1/2}$ = 260(10) ms [1]) fit well with the systematic trend of the heavier U isotopes, providing further support for the assignment made above. Figure 2(b) shows a plot of hindrance factors for U and Pu isotopes, defined as the ratio of the g.s. to g.s. to g.s. to 2^+ α decay reduced widths. The α decay reduced widths were calculated using the formulism of Rasmussen [15]. The hindrance factors for the isotopes of U are parabolic around neutron number 138. Again the value obtained in this work (HF = 0.65(9)) fits well with the systematic trend.

3.2 Alpha Decay of ^{230}Pu

In the case of ^{230}Pu, the events of interest were selected by searching for position and time correlated pairs of α-α events. Here, the second event had the constraint of having

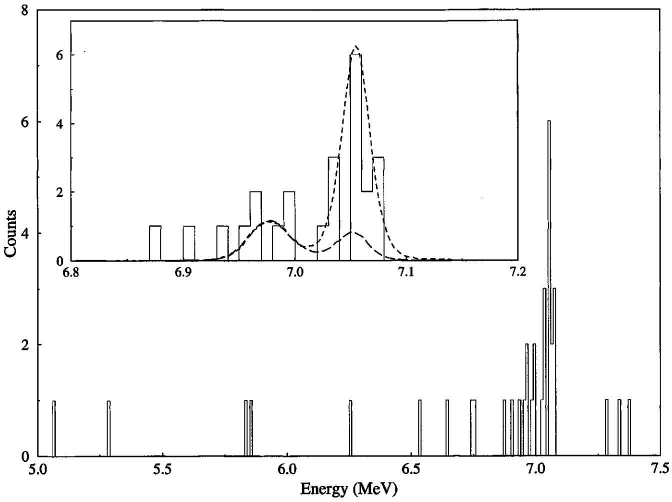

Fig. 3. Events which are position and time correlated with the α decay of either ^{226}U or ^{222}Th, within a maximum search interval of 900 ms. The inset to this figure shows an expansion of the region around 7.0 MeV. The dotted and dashed lines correspond to a Monte Carlo simulation; see text for details

an energy between (7545 to 7585) or (7960 to 8000) keV corresponding to the known g.s. to g.s. α-particle energies of ^{226}U and ^{222}Th, respectively. The maximum allowed time interval between pairs of events was 900 ms. Again the vertical position window was approximately $\pm$ 1 mm. The spectrum obtained is shown in Fig. 3.

It can be seen that there is a distribution of events at lower energy than that of the main peak. The energy of the main peak is 7055(17) keV, consistent with that measured for the α decay of ^{230}Pu [16]. The measured centroid of the distribution of events at lower energy is 6970(19) keV. The measured intensity ratios are 68(24)% and 32(16)% for the higher and lower energy distributions, respectively. These values are similar to the α-decay branching ratios observed in the heavier Pu isotopes [14]. Thus it is reasonable to assume that the lower energy distribution corresponds to the α decay of ^{230}Pu to the 2^+ state of ^{226}U. However, if this is the case, it is expected that the measured centroid will be shifted to higher energy, again due to the effect of conversion electrons summing with the α-particle energy. In this case, since the transition energy is not known, the energy of the 2^+ to 0^+ transition input into the Monte Carlo code is varied, and the centroid separation of the two peaks in the resulting spectrum measured, in order to obtain a calibration curve. The output from the simulation which best reproduces the experimentally measured centroid separation is shown as the dotted line in the inset of Fig. 3. The results of this procedure show that the 2^+ to 0^+ transition energy in ^{226}U is 96(25) keV, consistent with the candidate observed in the RDT experiment. Thus the α-particle energy of the decay ^{230}Pu (g.s.) to ^{226}U (2^+) is 6961(30) keV, again taking into account the small difference recoil energy terms. As in the case of the decay of ^{226}U, the measured intensity ratios must be corrected in order to obtain the branching ratio. In this case, the fraction of events that are shifted into the g.s. to g.s. peak is 35%. The corrected branching ratios are then 51(22)% and 49(25)% for the decays to the ground and 2^+ states, respectively.

The measurement of the 2^+ to 0^+ transition energy allows an estimate of the deformation parameter β_2 to be made, using the empirical relationships of [17,18]. The deduced value for ^{226}U, using 96 keV, is $\beta_2 \simeq 0.20$. This is similar to that of the isotone, ^{224}Th, and follows the systematic trend of decreasing deformation with decreasing mass number observed in the heavier U isotopes. The measured half-life for the 7565 keV ^{226}U α decay events was 230^{+60}_{-40} ms, obtained using the maximum likelihood method described in [19]. This value is in good agreement with previously published values (260(10) ms and 200(50) ms from [1,10], respectively). The value of the ^{226}U α-decay half-life obtained here, along with that published in [1], are however inconsistent with the somewhat lower value of 160^{+21}_{-17} ms published in [11]. A recent re-evaluation of the latter data, however, produced a value of 250(40) ms showing that the half-lives obtained from all data sets are consistent [20]. Using the Geiger-Nuttall relationship between α-decay Q-value and partial half-life, and the decay characteristics of the heavier Pu isotopes (taken from [14]), it is possible to estimate the half-life of ^{230}Pu. The partial half-life for the g.s. to g.s. ^{230}Pu α-decay line observed in this work is deduced from an extrapolation of the Geiger-Nuttall curve for the Pu isotopes. The resulting curves are shown in Fig. 2(a). The deduced α half-life of ^{230}Pu using the branching ratios quoted above is 154(66) s. This is much longer than the average time interval between events in the position elements of the Si-strip detector, and so it was not possible to measure the ^{230}Pu α half-life in this experiment. Assuming that the partial half-life for decay by electron capture is 800 s [21], and using the present data, the branching ratios for electron capture and α decay are estimated to be 16% and 84%, respectively.

Using the extrapolated value for α half-life together with the deduced branching ratios, the hindrance factor for the decay of ^{230}Pu has been calculated. The value obtained, HF=0.27(24), is plotted along with those for the heavier Pu isotopes in Fig. 2(b). Within the error bars, it appears that the hindrance factors for the Pu isotopes continue to decrease below neutron number 138. An improved measurement of the branching ratio along with a determination of the decay half-life would be desirable in order to confirm this trend.

4 Summary

Fine structure in the α decay of ^{230}Pu has been observed for the first time. In addition, improved measurements of the α-particle energies and branching ratios from ^{226}U have been obtained. The excitation energy of the first excited 2^+ state in ^{226}U has been determined to be 96(25) keV, giving an estimate of the deformation parameter, β_2, of 0.20, which follows the systematic trend of decreasing deformation with decreasing mass number observed in the heavier U isotopes. The half-life for the α

decay of ^{230}Pu is estimated to be approximately 154(66) s, from an extrapolation of the Geiger-Nuttall curves for the heavier Pu isotopes. The calculated hindrance factors are parabolic around neutron number 138 for the U isotopes, whilst for the Pu isotopes, it appears that the hindrance factor continues to decrease with decreasing neutron number.

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