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Spectroscopy of actinide nuclei – Perspectives with position sensitive HPGe detectors

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Abstract

Recent advances in in-beam gamma-ray spectroscopy of actinide nuclei are based on highly efficient arrays of escape-suppressed spectrometers. The sensitivity of these detector arrays is greatly enhanced by the combination with powerful mass separators or particle detector systems. This technique is demonstrated by an experiment to investigate excited states in $^{234}$U after the one-neutron-transfer reaction $^{235}$U(d,t). In coincidence with the outgoing tritons, $\gamma$-rays were detected with the highly efficient MINIBALL spectrometer. In the near future an even enhanced sensitivity will be achieved by utilizing position sensitive HPGe detectors which will exploit the novel detection method of gamma-ray energy tracking in electrically segmented germanium detectors. An example for this novel approach is the investigation neutron-rich actinide Th and U nuclei after multi nucleon transfer reactions employing the AGATA demonstrator and PRISMA setup at LNL, Italy. A primary $^{136}$Xe beam hitting a $^{238}$U target was used to produce the nuclei of interest. Beam-like reaction products after neutron transfer were selected by the PRISMA spectrometer. Coincident $\gamma$-rays from excited states in beam and target like particles were measured with the position sensitive AGATA HPGe detectors. Improved Doppler correction and quality of the $\gamma$-spectra is based on the novel $\gamma$-ray tracking technique, which was successfully exploited in this region.

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1. Introduction

Spectroscopic studies of nuclei in the actinide region progressed in recent years due to the increased sensitivity of highly efficient gamma-ray spectrometers and their combination with powerful devices for particle detection like mass separators or charged particle detector arrays. In this way the knowledge of the nuclear structure of these heavy nuclei was considerably extended (Leino and Heßberger, 2004; Herzberg and Greenless, 2008). Excited states in the first but also of the highly deformed second minimum of the double humped fission barrier were accessible (Thirolf and Habs, 2002; Pansegrau et al., 2000). The actinide nuclei, which are available in sufficient amount for target production, allow for detailed investigations by means of Coulomb excitation and close by nuclei are reached via transfer reactions.

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For even heavier nuclei in the transfermium region with \( Z \geq 100 \) the ‘cold fusion’, compound nucleus reactions were successfully employed for in-beam \( \gamma \)-ray spectroscopy from fermium (Bastin et al., 2006) up to rutherfordium (Greenless et al., 2012). Especially in these experiments the overwhelming background for prompt \( \gamma \)-ray detection at the target position from excited states of fission fragments was successfully coped with by adapting successfully the Recoil Decay Tagging (RDT) technique (Leino and Heßberger, 2004)). Moreover the refined experimental techniques allow for detection of long sought isomeric states in these barely known nuclei. The high efficiency of the spectrometers, coming from nearly complete solid angle coverage, together with the high detector granularity enables the measurement of the excitation energy and the angular moment after neutron evaporation in the surviving evaporation residue (Reiter et al., 2000). With these new observables the saddle point energies, fission barriers and shell correction energies are accessible and allow new insights into the reaction mechanism (Henning, 2012).

In the near future the field will profit from a new generation of position sensitive High Purity (HP) Ge-detectors which will allow improvements of the basic requirements for spectroscopic studies in this mass region. The highest efficiency can be obtained by using Ge detectors without escape-suppression shields or Anti-Compton shields, which surround the germanium by BGO scintillator detectors. Although the escape suppression significantly improves the quality of the spectra, the shields occupy a significant fraction of the 4\( \pi \) solid angle which reduces the overall efficiency of \( \gamma \)-ray detection. Therefore a new generation of gamma-ray spectrometers is based on HPGe detectors without physical suppression shields. In this way the ultimate goal of a 4\( \pi \) germanium shell is feasible. However the new spectrometer has to include the novel technique of gamma-ray energy tracking in electrically segmented germanium detectors. The gamma-ray tracking spectrometer will have increased detection sensitivity for selection of weakest decay channels from surviving heavy nuclei in the presence of background radiation from the overwhelming fission radiation (Eberth and Simpson, 2008).

A gamma-ray tracking system involves measuring the position and energy of every gamma-ray interaction in a detector so that the path and sequential energy-loss of a single gamma ray can be deduced using the Compton-scattering formula. The full energy of the event can then be reconstructed without the losses due to suppression shields, which covered nearly half the solid angle in the previous generation of spectrometers. The realization of such a system requires highly segmented germanium detectors and digital electronics to extract energy, time, and position information using pulse-shape information. The AGATA demonstrator is such an advanced gamma-ray tracking system and started operation at the Legnaro National Laboratories (LNL) in Italy (Akkoyun et al., 2012).

In this contribution two examples for in-beam \( \gamma \)-ray spectroscopy with advanced spectrometers will be presented. First the results of an experiment to study \( ^{234}\text{U} \), performed with the MINIBALL spectrometer (Warr et al., 2013) at the tandem accelerator of the University of Cologne will be discussed (Kotthaus et al., in press). Second the feasibility of spectroscopic investigations in the actinide region with the AGATA demonstrator and the novel gamma-ray tracking technique will be shown.

2. Spectroscopy of \( ^{234}\text{U} \)

Nuclei in the actinide region are distant from shell closures, have quadrupole-deformed shapes and exhibit well-developed rotational band spectra. Below the pairing gap at 1.5-2 MeV shape oscillations in the quadrupole and the octupole degree of freedom give rise to several low-lying rotational bands, namely the band with \( K=2^+ \) and octupole vibrations. There is usually also a low-lying band with \( K=0^+ \) present in this region, which might be the \( \beta \)-vibration or another kind of collective excitation (Garret, 2001). In addition to these one phonon excitations the collective nuclear model by Bohr and Mottelson (Bohr and Mottelson, 1975) predicts two and more-phonon excitations at higher energies. However, in contrast to spherical nuclei, where multi-phonon excitations are quite common, there have been only few reports of observations of unambiguous two-phonon excitations in deformed nuclei, mostly in the region of the rare earth isotopes (Börner et al., 1991; Corminboeuf et al., 1997; Fahlander et al., 1996; Oshima et al., 1995). In the actinide region only the nucleus \( ^{232}\text{Th} \) shows an \( 4^+ \) state fulfilling the requirements of a double phonon excitation (Korten et al., 1993; Korten et al., 1995). Therefore, a spectroscopic study of \( ^{234}\text{U} \), the neighbouring heavier even-even isotope of \( ^{232}\text{Th} \) was performed addressing the question whether or not multi-phonon excitations exist as common feature in deformed heavy nuclei.

New results were obtained by measuring the transitions following the one-neutron transfer reaction \( ^{235}\text{U}(d,p)^{234}\text{U} \) with the high-resolution 4\( \pi \) spectrometer MINIBALL (Warr et al., 2013). The reaction channel is selected by identification of the light charged reaction product and \( \gamma\gamma \) coincidence data is recorded. The spectrum from the new in-beam \( \gamma \)-ray spectroscopy experiment is shown in Fig. 1. It enabled the derivation of an extended level scheme of \( ^{234}\text{U} \). In comparison with previous investigations in the actinide region, the high efficiency of the MINIBALL \( \gamma \)-ray spectrometer allowed the analysis of coincidence data, which was not feasible in a previous \( \gamma \)-ray measurement (Ardisson et al., 1986). The analysis of the coincidence data allows for several extensions and corrections of the known level scheme of \( ^{234}\text{U} \). Strong transitions
between a $4^+$ state at approximately twice the energy of the $\gamma$-vibration of 1886.7 keV and it features strong decays to the rotational band built on the $\gamma$-vibration. Both observations are necessary for an expected double-phonon state.

![Image](image.png)

Fig. 1. The $\gamma$-spectrum of $^{234}$U: transition energies, which are assigned to the level scheme of $^{234}$U are marked by a black dot. The close-lying multiplets were resolved by analyzing the coincidence data.

These findings are compared to the neighbouring isotope $^{232}$Th. Here the state in question is identified as a two-phonon state also via lifetime measurements and absolute $B(E2)$ values (Korten et al., 1995). In $^{234}$U, the lifetime of the state and absolute $B(E2)$ values of the interesting transitions are not known. The strong population of this $4^+$ state via the one-neutron transfer reaction $^{235}$U(d,t) is at a first glance not in line with the two-phonon state interpretation. Consequently, the quest for a two-phonon state in $^{234}$U cannot be decided on the basis of the existing data. In order to identify the collective character of the discussed candidate of the two-phonon state complementary experiments in $^{234}$U are of highest interest. The answer to the question whether or not multi-phonon excitations exist as common feature in deformed heavy nuclei will remain open and will be subject of future experiments.

3. Spectroscopy of neutron-rich actinide nuclei

The region of neutron rich actinides is hardly accessible for experiments probing the nuclear structure. One way to perform gamma ray spectroscopy in the region of neutron-rich Thorium to Plutonium nuclei is based on the production of the interesting nuclei via multi-nucleon transfer reactions. In our case a $^{136}$Xe beam with a total energy of 930 MeV was directed on a $^{238}$U target. The beam was provided by the Tandem-PIAVE-ALPI accelerator complex at the LNL, Italy. The beam-like particles were identified using the PRISMA magnetic spectrometer (Corradi et al., 2009) at a rotation of 50°. The $\gamma$-rays were measured by the AGATA demonstrator (Akkoyun et al., 2012) facing the entrance window of PRISMA. Furthermore, position sensitive multi-channel-plate detectors served for particle identification. They were located inside the scattering angle to detect the recoiling target-like particles in order to allow measurements of particle-particle coincidences. A schematic picture of the set-up and additional information on the experimental details are given in Fig. 2.

The PRISMA spectrometer measures the velocity vector of the identified ejectile. This information is used to calculate the velocity vector of the target like particles by requiring momentum and energy conservation. In combination with the high
position resolution of the AGATA spectrometer and the possibility to identify the first interaction of emitted γ-rays from excited reaction products the Doppler correction of the γ-rays can be performed in two ways for the two outgoing reaction products.

Five different elements, namely Xenon (100%), Iodine (37%), Cesium (33%), Tellurium (19%) and Barium (16%), were identified after the reaction by looking at the energy loss in the ionization chamber of PRISMA. Many different isotopes for each element are found; for Xenon the chain from $^{126}\text{Xe}$ up to $^{144}\text{Xe}$ is identified. The measurement of mass and charge of the reaction product is based on the time-of-flight of the particle in combination with the calculation of its trajectory through the full PRISMA detector, especially through the large dipole magnet. The results yield an A over Q ratio providing A the mass and Q the charge of the particle. The radius of the trajectory in combination with the velocity and the full energy deposited in the ionization chamber allows identification of the ionic charge state of the particle and therewith the different isotopes. The analysis of the target like reaction products is based on the assumption that by gating on a specific ejectile isotope the corresponding isotope of the recoil is produced. For example, in case $^{134}\text{Xe}$ is identified as the two-neutron transfer channel ($^{136}\text{Xe}-2\text{n}$) then the corresponding actinide nucleus $^{240}\text{U}$ is produced as well. However, the excitation energy of the heavier reaction product may in several cases exceed the fission barrier or the neutron binding energy and the corresponding final exit channel will be fission fragments or neutron evaporation residues.

Fig. 2. Schematic drawing of the experimental set-up of AGATA demonstrator and PRISMA spectrometer at LNL used for the measurement of a primary $^{136}\text{Xe}$ beam at an energy of 1 GeV hitting a backed $^{238}\text{U}$ target. The nuclei of interest are produced via multi-nucleon reactions. The PRISMA spectrometer detects beam-like reaction products. Surviving, highly fissile actinide reaction product are also detected with the position sensitive multi-channel plate detector of the DANTE array which was positioned inside the scattering chamber in the reaction plane covering the angle range which corresponds to the grazing angle for the target like reaction product.

The recorded γ ray spectra are Doppler corrected for beam and for the target like nuclei. In order to obtain these spectra the pulse shape information of the detector signals of the AGATA demonstrator was recorded and a pulse shape analysis (PSA) is performed to identify the position of each interaction within a segment of each detector. The PSA algorithm compares the recorded signals with a database containing a huge library of calculated data sets for all positions inside the large volume HPGe detectors on a 3 dimensional grid with a 2 mm grid size. An intermediate analysis step corrects the energy for cross talk and for charge carrier trapping caused by neutron damage of the detector. Then the deduced position information from all interaction points inside the detectors is processed by a gamma ray tracking (GRT) algorithm. The GRT code combines the position information from all 15 detectors AGATA demonstrator detectors and identifies the three main interaction processes (photo effect, Compton scattering or pair creation) within the complete demonstrator array. Moreover the first interaction position of the individual γ-ray is obtained on an event-by-event basis. This information is combined with the information on the velocity vector coming from the PRISMA separator to create the Doppler corrected spectra. In this way only the position resolution of the new detectors and the uncertainty in velocity will finally determine the quality of the Doppler correction. The Doppler broadening will be clearly reduced in comparison with standard HPGe detectors. In this case the considerable opening angle of the large volume detector determines the width of the γ ray line. Furthermore a gate on the prompt time peak is crucial for additional background reduction between.
As a first result clean γ-ray spectra are obtained for the individual isotopes. In Fig. 3 the individual peaks from transitions of the different Xe isotopes are visible in background free spectra. The corresponding results for the Uranium partners are in a preliminary stage and subject of the ongoing data analysis. In order to illustrate the capability of the γ-ray tracking procedure the results from the ground state band in $^{238}$U is shown in Fig. 4. The four overlaid spectra illustrate the improvements at various steps of the elaborate analysis. The core signal spectrum (green) reflects the line shape and energy resolution of the individual large volume HP Ge detectors and the considerable broadening already at low γ-ray energies. At this point no γ-ray tracking and no Doppler correction are performed. The γ-spectra are also shown after applying the tracking algorithm without (black) and with (red) Doppler correction. An improved peak-to-total ratio in spectrum (blue) is obtained by gating on the two outgoing particles. A proper coincidence condition enhances the peak-to-total ratio and at this point the spectrum quality of the new γ-ray tracking array is equivalent with the results from an array of large volume Compton shielded HPGe detectors. Additional analysis will allow a detailed investigation of other uranium nuclei and isotopes from the other elements (Birkenbach, 2014)).
4. Conclusion

After tremendous progress in spectroscopic studies of highly fissile actinide nuclei the field will benefit in future from position sensitive HPGe detectors and the novel $\gamma$-ray tracking technique. This will allow a higher sensitivity, efficiency and count rate capability for in-beam studies in this mass region and will overcome the existing limitations.

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