EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

Letter of Intent to the ISOLDE and Neutron Time-of-Flight Committee

Are ^{221}Rn and ^{223}Rn good candidates to search for an atomic EDM? Investigation of their low-energy nuclear level schemes following β decay of ^{221}At and ^{223}At

May 29, 2014

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CERN-INTC-2014-042 / INTC-I-158
29/05/2014

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Abstract: Our collaboration intends to clarify the question whether some atoms containing odd-mass Rn nuclei near A≈224 are suitable candidates in the search for $\mathcal{CP}$-violating static electric dipole moments. These will be enhanced by an octupole deformation of the nucleus, which manifests as parity doublets in the low-lying level schemes of these odd-mass isotopes. In order to test the existence of parity doublets, we propose to measure ^{221}Rn and ^{223}Rn following the β decay of ^{221}At and ^{223}At exploiting the capability of ISOLDE to provide comparably pure beams of these rare At isotopes. For the measurement we will employ the new ISOLDE Decay Station, which will allow for the measurement of transition energies, intensities and multipolarities. The primary aim is to determine the excitation energy of the first excited negative-parity state in ^{221}Rn. In addition, this measurement will complement a previous Coulomb-excitation measurement of ^{221}Rn at REX-ISOLDE and serve to provide information on whether the beam intensities and purities are suitable for lifetime measurements employing the fast-timing technique. However, at present it is not clear whether strong contamination from Francium and Radium isotopes will prevent a successful measurement. We estimate that in order to fulfil our physics aims we require a ratio of less than 100 contaminating nuclei to one nucleus of interest.

1 Physics case

Nuclei with certain nucleon numbers (N = Z = 34, 56, 88, 134) are known to exhibit features associated with an enhanced octupole collectivity [1]. The shell structure of these nuclei is characterized in such a way that the Fermi-level is situated between the unique parity subshell and the subshell differing by three units in total ($\Delta j = 3$) and orbital ($\Delta l = 3$) angular momentum. A coherent superposition of particle-hole excitations involving this particular $[nl_j, (n-1)(l+3)_{(j+3)}]_{3^-}$ subshell combination leads to enhanced octupole collectivity. In the mass region of interest these subshell combinations are the proton $[2f_{7/2}, 1i_{13/2}]$ and neutron $[2g_{9/2}, 1j_{15/2}]$ configurations. Octupole collectivity is particularly pronounced in nuclei for which such a shell structure is realised for protons as well as for neutrons. If such a configuration is present, the residual long-ranged proton-neutron interaction drives the nucleus to adopt a pear shape.

For even-even nuclei the major experimental feature associated with this reflection asymmetric shape is the staggering of interleaved positive- and negative-parity yrast levels. In the actinide region corresponding level sequences (see Fig. 1(b)) were observed in multi-nucleon transfer reactions [2]. Furthermore, these nuclei necessarily possess low-lying $J^\pi = 3^-$ states, which are expected to be connected to the ground state with a strong $B(E3)$ transition probability. Indeed, a recent measurement of the latter quantity at REX-ISOLDE [3] confirmed for ^{224}Ra the presence of octupole correlations in its ground state, which are associated with a permanent octupole deformation. For ^{220}Rn, results of the same campaign led to the conclusion that the octupole collectivity in this nucleus is of vibrational nature.
A static octupole moment result due to the interplay with the quadrupole deformation in a $\mathcal{CP}$-violating nuclear Schiff moment. Yet, the considerations with respect to $\mathcal{CP}$ violation demand the presence of a predominant direction which is in an odd-mass nucleus defined by the projection of an intrinsic angular momentum on the symmetry axis. The K-quantum number projection and the Schiff moment form a pseudo scalar which is no longer invariant under the $\mathcal{CP}$ operation. Furthermore, in these nuclei the octupolarity leads to almost degenerate parity doublets consisting of two states with the same angular momentum but opposite parity. The latter is a direct consequence of the $\mathcal{RPT}$ symmetry for a reflection asymmetric quantum system with $K \neq 0$ [4] assigning to the positive and negative yrast levels the same simplex quantum number. In case the nuclear force contains a $\mathcal{P}$-($\mathcal{T}$-)odd part the levels forming the parity doublet can mix. In the lowest order this mixing itself is determined by the octupole deformation (β_3) but enhanced by the intrinsic Schiff moment ($\propto \beta_2 \cdot \beta_3$). In the laboratory frame [5, 6, 7] the resulting Schiff moment is proportional to:

$$\propto \frac{\beta_2(\beta_3)^2}{\Delta E_\pm}, \tag{1}$$

where $\Delta E_\pm$ is the energy difference between positive- and negative-parity member of the parity doublet. Due to the small energy spacing between the parity doublets and the relatively large matrix elements connecting (via a $\mathcal{P}$-odd, $\mathcal{T}$-odd interaction) the doublet states, the intrinsic nuclear Schiff moment is enhanced [5, 6, 7]. Clearly, because of the weakness of the $\mathcal{P}$-odd, $\mathcal{T}$-odd part of the nuclear force it is, in the foreseeable future, not feasible to measure this nuclear Schiff moment. Nevertheless, in the atomic system the interaction of the electrons with the associated asymmetric nuclear charge distribution will not only transfer to but even amplify the Schiff moment in the atomic system [8, 9]. In fact, theory predicts for atoms containing an octupole deformed nucleus an enhancement of the atomic electric dipole moment (EDM) by a factor of 1000 in comparison to ^{199}Hg, which currently sets the experimental lower limit for an atomic EDM ($d(^{199}\mathrm{Hg}) < 3.1 \times 10^{-29}$ e·cm) [10].
The energy denominator entering Equation 1 is a crucial quantity and, from a spectroscopist's point of view, the easiest accessible observable. The presence of such parity doublets is well established in odd-mass Ra and Th isotopes such as, for example, ^{223}Ra (see Fig. 1 in Reference [12]), ^{225}Ra, and ^{227}Th [13]. Owed to the fact of Radon being a noble gas, it has favourable chemical properties supporting the search for an atomic EDM. Therefore, it is of high interest whether the structure of any of its nuclei enhances the atomic EDM due to the pear shape associated with the interplay of quadrupole and octupole deformation. The previous statement that the octupole collectivity in ^{220}Rn is vibrational would, in principle, rule out the observation of such parity doublets in ^{221}Rn. Nevertheless, at present no data concerning the B(E3, $0^+ \to 3^-$) strength for the other possible core ^{222}Rn is available. Furthermore, theory [14, 15] emphasises for odd-mass nuclei the possibility that core polarisation induced by the interaction with the odd particle results in an octupole-deformed shape.

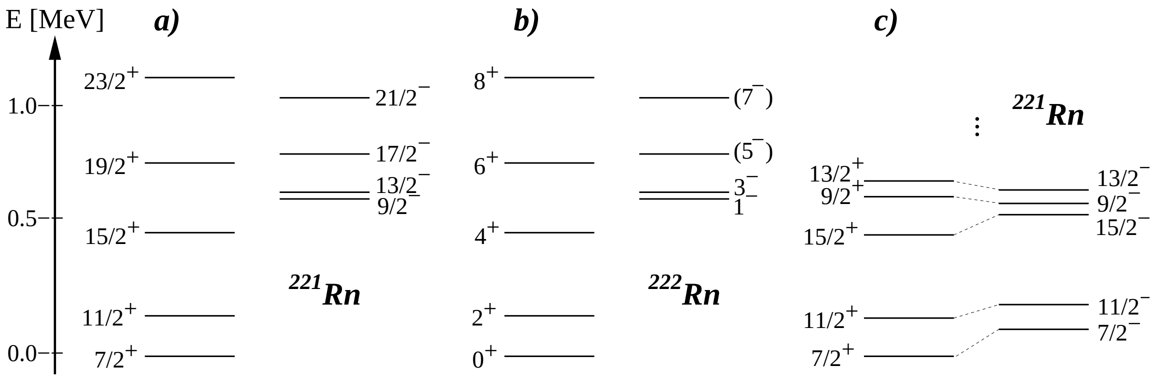

Figure 1: Schematic low-lying level schemes of ^{221}Rn applying the strong coupling limit without octupole deformation (part a) and with octupole deformation (part c) based on the known levels of a ^{222}Rn [2] core (part b). For clarity, possible single-particle states other than the $J^\pi = 7/2^+$ ground state were neglected. In part c the parity doublets, which are characteristic for the presence of an octupole deformation in an odd-mass nucleus, are indicated by the dashed lines connecting the corresponding levels.

The low-energy level scheme of the ^{222}Rn core [2] is shown in the middle part of Figure 1. The low excitation energy of the first excited 2^+ level already suggests an amount of quadrupole deformation in the ground state of this isotope. This is supported by the fact that the band head of the octupole band is given by a 1^- level originating from the interplay of the octupole excitation and the ground-state quadrupole deformation.
Due to the already present quadrupole correlations, the coupling of the unpaired particle to the even-even core can be expected to fulfil the conditions of the strong coupling limit [11]. In this coupling scheme the particle-core coupled states of the odd-mass nucleus are expected at the same excitation energy as observed for the corresponding even-even core nucleus. However, in presence of parity doublets as signature of an octupolar system the odd-mass nucleus would also have negative-parity states at much lower energies than expected, following the strong-coupling limit. The resulting level schemes for a strong coupling limit without octupole deformation is shown in Fig. 1a and with octupole deformation in Fig. 1c. In order to assure clarity of the presentation, in Fig. 1 other single-particle levels and their couplings were neglected.
Since most theoretical studies in the mass region of interest concentrate on the Ra and Th isotopes near $N = 134$, theoretical studies for the isotopes of interest are rather sparse. These investigations suffer from the transitional nature of these nuclei. Consequently, studies using the Quasiparticle Random Phase Approximation (QRPA) (e.g., see Ref. [7]) predict a rather vibrational character as limited by the applicability of RPA based models. Models using reflection-asymmetric rotor models assume a well pronounced deformation (e.g., see Refs. [15] and [16]). Interestingly, the latter approach is capable of reproducing for the experimentally observed ground-state properties (magnetic and quadrupole moment) of ^{223}Rn by mixing various Ω^+ Nilsson orbits ($\Omega^\pi = 3/2^+$ and $\Omega^\pi = 1/2^+$); but fails for ^{221}Rn. The authors of Reference [16] explicitly demand more spectroscopic data of excited levels.
The presently available spectroscopic data on excited states in the isotopes of interest is limited to one tentatively assigned excited state in ^{221}Rn at ≈ 30 keV measured following α decay of ^{225}Ra [17]. Furthermore, in a first Coulomb-excitation experiment at REX-ISOLDE, several γ rays associated with ^{221}Rn were observed. Owing to insufficient statistics, it was not possible to create a $\gamma\gamma$ matrix and firmly assign these γ rays to a level scheme. For ^{223}Rn, no information about any excited levels exists at all.

For ^{221}At and ^{223}At only limited spectroscopic data are available, for example, not even their ground-state spins are known. The lack of data is also highlighted by the imprecise knowledge of the half-lifes (^{221}At: 2.3(2) min; ^{223}At: 50(7) s). For both isotopes the β-decay branch is the dominating decay process ($B_\alpha \leq 0.001$ %) [18]. The β-decay Q-values, as given in the AME2012 atomic mass evaluation compilation [19] amount to ^{221}At: 2310(15) keV and ^{223}At: 3038(16) keV. Basically, the Q-values for the two mother isotopes are sufficient to populate low-lying levels in the daughter isotopes.

2 Beam considerations for ^{221,223}At β-decay experiments

Recently, the first determination of the Astatine ionisation potential has received a great deal of attention [20]. This advance allows for a selective laser ionisation exploiting RILIS which results, for the neutron-deficient At isotopes, in pure beams. However, for the isotopes of interest the primary yields for the alkali (Fr) and earth-alkali (Ra) isobars are immense (e.g., ^{221}Fr measured at the SC: 8.9×10^8Ions/μC (ThC_X) or 2.8×10^7Ions/μC (UC_X) [21]). These enormous production yields in combination with their low ionisation potential results in considerable beam contamination, which might exceed the isotopes of interest by orders of magnitude. Indeed, previous experiments using neutron rich At beams up to ^{219}At have shown an onset of strong beam contamination consisting of various Francium isotopes when approaching the isotopes of interest [22]. The enormous production yields for the isobaric contaminants out-weight the production yields for the isotopes of interest (^{221}At SC: 4500 ions/μC (ThO_2); ^{223}At SC: 100 ions/μC ($Th0_2$) [21]) by far. Therefore, at present, a successful β-decay measurement of ^{221}At and ^{223}At is doubtful. Attempts at TRIUMF failed and in the next two years no further opportunity is given [23]. At ISOLDE beam development for the isotopes of interest has already been requested in earlier Letters of Intent [24, 25] by members of our collaboration.
In order to ensure comparably pure beams of the mother isotopes, ^{221}At and ^{223}At, we request the development of these beam. These development contains the adjustment of the At laser-ionization scheme to the isotopes of interest, as well as the test of methods to suppress the disturbing Fr and Ra isotopes. The latter methods are a quartz transfer line and if necessary a combination of quartz transfer line and the Laser IoniSation Trap (LIST). In any case the use of the High-Resolution Separator (HRS) is mandatory. The measurement itself is intended to employ the ISOLDE Decay Station (IDS), which is currently under construction.
The issues arising from the contaminants are twofold. First of all the additional decays will raise the dead time in the detectors and electronics. This issue can be counteracted either by reducing the proton beam on the primary target or a more narrow setting of the slits in the HRS. In any case, the additional decays will contaminate the obtained $\gamma\gamma$ and γe^- matrices. We intent to use a laser on/off approach to obtain in a subtraction procedure $\gamma\gamma$ and γe^- matrices containing exclusively transitions associated with Rn daughter nuclei. In order to keep background fluctuations resulting from the subtraction procedure to be acceptable, we estimate that the ratio of recorded contaminant to isotope of interest events should not exceed 10:1.
Considering their by more than an order of magnitude different half-lives, this 10:1 condition results for ^{223}At ($T_{1/2} = 50$ s) with its β-decaying isobar ^{223}Fr ($T_{1/2} = 22$ min) in an acceptable ratio of approximately 1:100. This ratio might lowered in case a considerable amount of ^{224}Fr ($T_{1/2} = 3.33$ min) is detected in the beam. Since, ^{222}Fr ($T_{1/2} = 14.2$ min) has a half-life in the same order of magnitude as ^{223}Fr the considerations as stated for this isotope are valid. Tuning the HRS appropriate will suppress the shorter lived ^{224}Fr and reduce the recorded activity

associated with the Fr isotopes. A critical point is the isobar ^{223}Ra. In spite that its half-life ($T_{1/2}$ = 11.43 d) and the low specific activity associated indicate it won't be an issue for this experiment, this α decaying isotope could, when reaching the IDS, contaminate the chamber for a longer period of time.
For ^{221}At the corresponding Fr isobar is more challenging. The half-lives of both isotopes are comparable (^{221}At: $T_{1/2}$ = 138 s; ^{221}Fr: $T_{1/2}$ = 286.1 s). Furthermore, ^{221}Fr decays almost exclusively via α emission. At a first glance this decay behaviour seems to be favourable because most α decays proceed to the ground state or to low-lying excited states from which they decay directly to the ground state. Consequently, apart from random coincidences these decays do not contribute to the recorded $\gamma\gamma$ or γe^- matrices. However, considering that the maximum energy to implant these nuclei into the Mylar tape of IDS is 60 kV and the recoil these nuclei experience in α decay is $\approx$ 130 keV, it can be estimated that $\approx$5 % of these nuclei are leaving the tape before their activity is transported away. Following the decay chain ^{221}Fr $\rightarrow^{217}$At $\rightarrow^{213}$Bi another β-unstable nucleus is reached. While the ^{223}Fr nuclei implanted in the tape won't contribute significantly to the overall count rates, the nuclei which are recoiled into the chamber will fully enter the recorded matrices. ^{220}Fr ($T_{1/2}$ = 27.4 s) is a short lived α emitter, which will lead to similar problems as encountered for ^{221}Fr while, as previously mentioned, ^{222}Fr is a comparably long-living β emitter. Consequently, an appropriate setting of the HRS might suppress ^{220}Fr and reduce the contaminants activity. The presence of the short-lived α emitter ^{221}Ra ($T_{1/2}$ = 28 s) in the beam is another source of contamination of the chamber and should be avoided. To summarize, for ^{221}At the ratio of desired to contaminating beam should not exceed 1:100.
Due to their comparable half-lives (^{221}Rn: $T_{1/2}$ = 25 min; ^{223}Rn: $T_{1/2}$ = 24.3 min), contributions to the accumulated activity by the decay of the Rn daughter nuclei can be neglected.

3 Summary

In summary, our collaboration requests further development of the neutron-rich Astatine beams at ISOLDE, the odd-mass isotopes ^{221,223}At being of particular importance to our physics interests of determining the excitation energy of the negative-parity states in their odd-mass Rn daughters and, therefore, testing the presence of octupole correlations in these nuclei. Key to a successful experiment is the demonstration that isobaric "purity" of 1:10 is achieved, although it is acceptable if it can be demonstrated to be better than 1:100.

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