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First results from laser spectroscopy on bunched radioactive beams from the JYFL ion-beam cooler

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Abstract. A new RFQ ion-beam cooler and buncher, installed after the mass-separating magnet of the ion guide isotope separator, IGISOL, JYFL, has dramatically increased the scope of on-line laser spectroscopy at this facility. The device, operated in a bunching mode, has permitted new measurements on short-lived radionuclei in the Ti, Zr and Hf chains at a sensitivity two orders of magnitude greater than that previously achieved. The device has also opened new prospects for laser-based nuclear spectroscopy at the facility, particularly collinear resonance ionisation spectroscopy.

PACS. 29.25.Rm Sources of radioactive nuclei – 32.10.Fn Fine and hyperfine structure

1 Introduction

Fundamental data on the structure of nuclear ground and isomeric states ($t_{1/2} \geq 1$ ms) can be derived from laser spectroscopic measurements in a model-independent manner [1]. While laser spectroscopy is competitive with several alternative techniques for the measurement of nuclear moments, it is unique in its ability to make precise comparisons of proton distributions between radioisotopes.

The laser spectroscopic station at the JYFL IGISOL (ion guide isotope separator) has now been enhanced by the addition of an on-line ion-beam cooler [2]. This device permits the rapid cooling and temporary ($\sim$ 200 ms) collection of radioactive ions produced by the IGISOL and greatly enhances the sensitivity of the subsequent optical spectroscopy. New measurements of the nuclear structural changes in the Ti, Zr and Hf isotope chains have been made using the device in an ion-bunching mode. In the continuous mode, access to low-emittance, low-energy spread, $\sim$ 0.6 eV, ionic ensembles can still be achieved without compromising the short release time ($\sim$ 1 ms) of the IGISOL production platform. This facility now provides the widest possible scope, in terms of elements, half-lives and sensitivity, for performing measurements across all regions of the nuclear landscape.

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2 Bunched-beam spectroscopy

A bunched-beam mode of ion source operation can, for sufficiently long-lived species, enable collinear ion-laser beam spectroscopy to be performed with an experimental sensitivity that compares to, or exceeds, all present forms of fast beam spectroscopy (including fluorescent ion tagging) [1]. The technique, at JYFL, is realised by applying an axial containment field to the cooler in order to trap the ions delivered by the IGISOL for up to 200 ms and, upon removing the containment field, subsequently releasing the ensemble in a 10–20 μs bunch. The re-accelerated ensemble (at 40 kV) is electrostatically formed to a spatially small, low-divergence beam and overlapped with a counterpropagating laser beam. A photomultiplier viewing an 18 mm region of this overlap is gated to register photons only as the ion bunch traverses the interaction region. The photon background in the fluorescent photon signal can be reduced in the ratio of the bunch width to accumulation time (10^{-4}).

We have now demonstrated, both off-line and on-line, the ability to cool, store and rapidly release ionic ensembles with little sensitivity to species chemistry or initial production beam properties (particularly the energy spread). Collinear spectroscopy has successfully been performed on samples at production rates of less than 100 ions per second.

Laser spectroscopic studies with bunched ion beams have been undertaken for three group-IVb isotope chains:

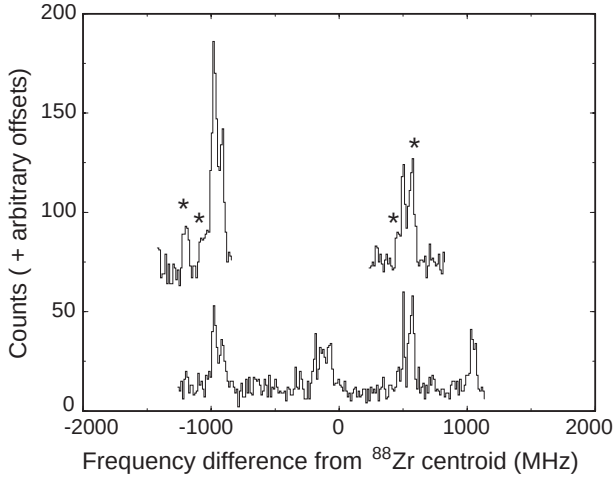

Fig. 1. Fluorescence spectrum of ^{87g,m}Zr with isomeric hyperfine components indicated by the stars.

Ti, Zr and Hf. Each system is very similar from an atomic perspective (with the ionic ground states all based on d^2s+ ds^2+d^3 configurations) and each has many ionic resonance lines in the 300–330 nm region. This region is accessible to frequency-doubled commercial dye lasers operating with the R6G or R640 laser dyes. Figure 1 shows a resonance fluorescence spectrum for ^{87g,m}Zr on the d^2s $^4F_{3/2}$-d^2p $^4D_{3/2}$ 310.0 nm line with an ion rate of $\sim$ 2000 ions per second for the ground state and $\sim$ 1000 ions per second for the isomeric state. The total accumulation time for the ground-state spectrum was 14 hours.

Although the systems of Ti, Zr and Hf have similar atomic structure, they exhibit very different nuclear structural properties. New measurements on these trends are summarised individually below. In each system the results on the radioactive isotopes represent the first reports of on-line measurements.

2.1 Neutron-deficient Ti isotopes

The only previous measurement of an isotope shift in radioactive titanium isotopes is that reported by Gangrsky *et al.* [3] for the long-lived ^{44}Ti isotope. The even-N Ti isotopes, down to $N = 24$, were reported to possess monotonically *increasing* charge radius with *decreasing* neutron number. Such a behaviour is distinctly different from that displayed by the Ca isotones, in which a well-known symmetric, parabolic charge radius trend is apparent (see fig. 2) in the $N = 20$–28 shell. The result for ^{44}Ti therefore appears to suggest a more surface-peaked or diffuser proton structure for Ti as the $N = 20$ shell closure is approached.

At JYFL, we have remeasured the ^{44}Ti charge radius and also report here a new result for ^{45}Ti. The Ti nuclei were produced using the ^{45}Sc(p, xn)$^{46-x}$Ti reaction at 15–25 MeV proton energy and studied on the ionic d^2s $^4F_{3/2}$-d^2p $^4F_{3/2}$ 324.2 nm line. Figure 2 shows the charge radii extracted from these measurements and compares them to their Ca isotones. It can be seen that the monotonic increase in the even-N radii is confirmed within errors

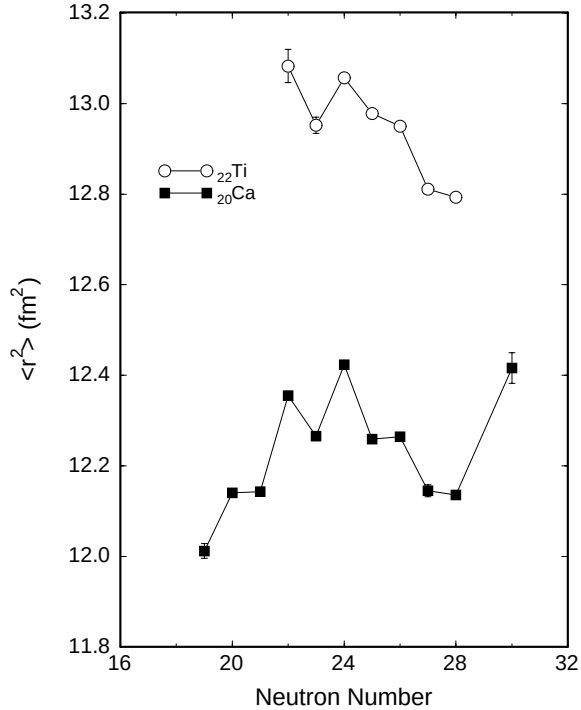

Fig. 2. The titanium and calcium mean-square charge radii (original references for the Ca data may be found in ref. [1]).

(consistent with ref. [3]). The decrease in charge radius observed at $N = 23$ (^{45}Ti) closely corresponds to that observed in Ca and only ^{44}Ti displays a substantial isotonic difference. The extension of these measurements to the $N = 20$ shell closure will be attempted at the IGISOL with ^{40}Ca(^{3}He, xn) reactions already used to study the properties of Ti nuclei across the shell closure [4].

2.2 Neutron-deficient Zr isotopes and isomers

The density and variety of nuclear shape changes that can be observed in the region around zirconium has resulted in the long-standing experimental interest in this region. However, with the exception of a low-accuracy measurement on the ^{88}Zr isotope, studied at the IGISOL prior to the installation of the cooler, no optical measurements have been made on the varied proton and neutron structures observed in radioisotopes in the $39 \leq Z \leq 46$ region.

Bunched-beam spectroscopy has now been performed on the neutron-deficient zirconium isotopes ($N \leq 50$). The isotopes were produced in the ^{89}Y(p, xn) reaction at 16–55 MeV with the ions studied on the d^2s $^4F_{3/2}$-d^2p $^4D_{3/2}$ 310.0 nm line. Figure 3 shows the new charge radii measurements for ^{87,87m,88,89,89m}Zr. The magnitude of the shell closure "kink" is comparable to those reported for the lower-Z isotope chains of Kr, Sr and Rb [1]. The departure from model estimates and trends suggested by the $B(E2)$ in this near-spherical region is large and also similar to that observed in the isotones [5]. For Kr, Sr and Rb, agreement between optical measurements, non-optical measurements and theoretical modelling is only achieved

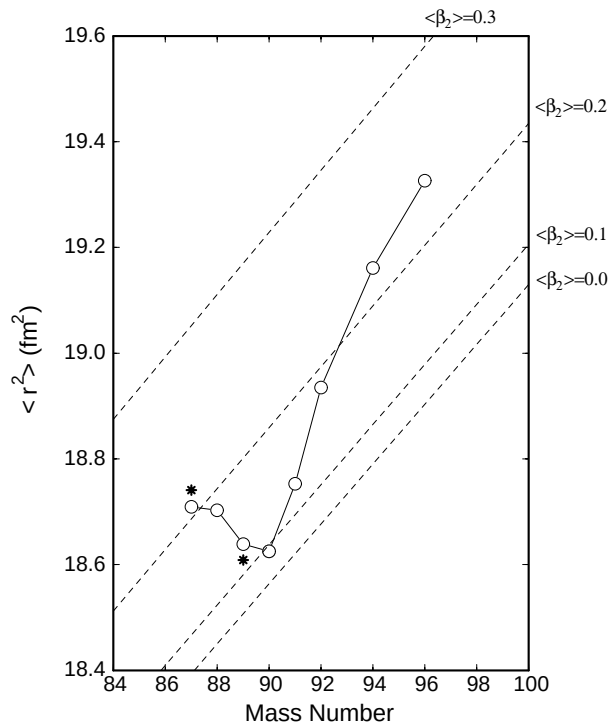

Fig. 3. The zirconium mean-square charge radii including the new measurements on radioactive isotopes and isomers (with isomers indicated by the asterisks).

for systems with strong collectivity and high deformations ($\beta_2 \sim 0.4$). Such deformations are found in the Zr isotopes, spectacularly, often within a few particle additions or subtractions (or indeed excitations) from near-spherical ground states. Studies of one region of such shape transition ($N \sim 58$) in Zr can be attempted at the IGISOL by use of proton-induced uranium fission. Explorations of these fission fragments are now underway.

2.3 The Hf isotopes and isomers

Investigations of the radial trends in the ground and isomeric states of radioactive hafnium commenced prior to the installation of the cooler [6]. With the cooler it has been possible to study the ^{175}Hf system even with reactions that produce background ion beams (target sputters) at the $A = 175$ mass. In the ion coincidence technique [6] such background beams would cause proportionate increases in the experimental background and could be highly detrimental to the spectroscopy. With bunched beams it has been possible to investigate the system using both the ^{175}Lu(d, 2n) and ^{176}Yb(α, 5n) reactions with the ion studied on the $ds^2\ ^2D_{3/2}$-$dsp\ ^2D_{5/2}$ 301.3 nm line.

A previous measurement of the ^{175}Hf charge radius [7] reported a surprisingly large normal odd-even staggering (OES) for this system (*i.e.* the charge radius of ^{175}Hf was less than the average of the two neighbouring even isotopes). The result is particularly surprising as other odd-N Hf isotopes in this region display a suppressed or inverted OES (similar to that of their higher-Z isotones) which arises from the large odd-N static quadrupole deformations.

The ^{175}Hf ground-state result reported here is found to be inconsistent with the previous report [7] and instead suggests that the ^{175}Hf ground state shows a large inverted OES and is in fact the most deformed ground state in the hafnium chain. The new charge radius measurement is now entirely compatible with the measured static quadrupole deformation in ^{175}Hf. A firm connection between the course of the radii and the intrinsic quadrupole, Q_0, moments (estimated from both lifetimes, $B(E2)$, and spectroscopic, Q_s, moments) has already been established across the rest of the chain [8]. The agreement between experiment and model predictions is also greatly improved and a comparison of the measured mean-square charge radii compared to droplet model predictions (corrected for the theoretical β_2 changes) is presented in fig. 4 [8].

Studies of the high-K isomers, abundant in the near-stable Hf region, will now be attempted at the IGISOL. The only presently existing measurement, ^{178m2}Hf [9], displays an interesting, large and negative isomer shift (fig. 4). This isomer is found to have a smaller charge radius than the ground state despite having a larger intrinsic quadrupole deformation. In this work we have shown that changes in charge radius in all other Hf states, including isomeric states, are almost exhaustively described by changes in size and static quadrupole deformation alone. For the ^{178m2}Hf system it may therefore be possible to investigate if the reduction in the isomeric radius, which cannot be described by shape and size changes, results instead from the reduction in nuclear pairing. Changes in

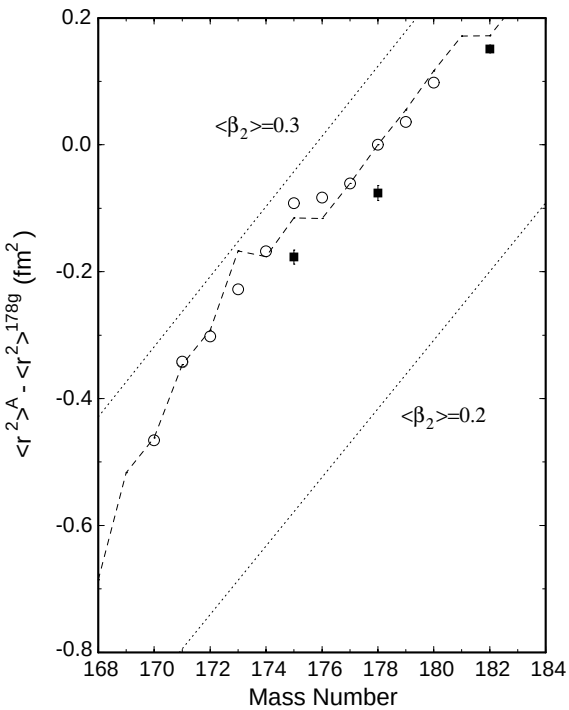

Fig. 4. The hafnium mean-square charge radii (with open circles indicating measurements reported here and squares indicating results from other works [9,10]).

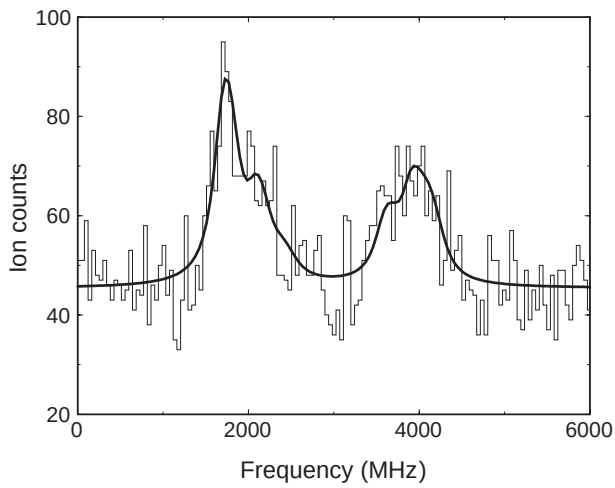

Fig. 5. Spectrum of ^{27}Al photo-ions against frequency. A Voigt profile fit to the compound structure is shown as a solid line.

nuclear pairing may reasonably be expected to result in changes in nuclear diffuseness (or nuclear rigidity) that can directly contribute to the mean-square value of the charge radius. The many high-K isomeric states in the near-stable Hf region provides the best possibility for attempting to resolve a role for the nuclear pairing in the course of the charge radius.

3 New developments

3.1 On-line stable beam production

Previously, in order to obtain data of sufficient resolution the IGISOL was run in an efficiency-compromised mode [6]. In this mode the skimmer electrode, which facilitates the collection of ions and the removal of the helium catcher gas, was operated at very low voltages (< 20 V). The cooler-buncher has removed this requirement and during our first on-line experience with high skimmer voltages a new feature was recognised. On-line, given sufficient voltage, a corona discharge is formed on the skimmer electrode, due to breakdown of the ionised He. If the skimmer plate is intentionally coated with a stable material, weak ion beams of that material are released with an energy corresponding to the potential of the skimmer electrode. Consequently, two beams of different average energy and different origin, one radioactive recoil and one stable skimmer coating, are produced on-line. The cooler energy acceptance window can be adjusted to select one of these beams for trapping and subsequent re-acceleration. In this manner it has been possible to move between radioactive measurements and calibrations with stable beams *on-line* with great speed and little disruption to the experiment. Great reductions in systematic measurement uncertainties have been achieved using such calibrations.

3.2 Collinear resonance ionisation spectroscopy

Collinear resonance ionisation is an extremely sensitive and proven spectroscopic tool [11]. Critically, however, the spectroscopy suffers from the low duty cycle of the high-power lasers if it is applied at continuous ion beam facilities. For typical Cu-vapour–based laser systems, with 10 kHz repetition rate, maximum duty cycles of only a few percent have been achieved [11]. At the IGISOL the cooler is capable of efficiently delivering bunched ion beams and it has been possible to perform high-efficiency, 100% duty cycle, collinear RIS using a 50 Hz Nd:YAG-based laser system.

Figure 5 shows a resonant photo-ion count for ^{27}Al taken against laser frequency in the region of the s^2p $^2P_{1/2}$-s^2d $^2D_{3/2}$ 308.2 nm transition. The fast Al^+ ions were neutralised via non-resonant charge exchange reactions with Na vapour. Two-step resonance ionisation was then performed using the amplified and doubled output of a Spectra Physics 380D dye laser (308 nm) and a brute force ionisation using $\sim$ 10 mJ of 532 nm Nd:YAG second harmonic. In its present configuration the cooler can produce bunches of 20 μs minimum duration, corresponding to $\sim$ 10 metres bunch length. Of this $\sim$ 1 m could be overlapped between charge exchange and ion detection (on a double stack of micro-channel plates). The spectrum in fig. 5 was taken with $\sim$ 20000 fast atoms per second of which $\sim$ 2000 per second were accessible for laser ionisation. The total accumulation time for the spectrum was 10 minutes. The increase in spectroscopic efficiency of more than two orders of magnitude (at the cost of a larger 300 MHz linewidth) may be seen by the comparison of the spectra in figs. 1 and 5 both taken at similar ion rates but with a factor of $\sim$ 80 difference in accumulation time.

4 Summary

The on-line ion beam cooler at the JYFL IGISOL is now fully operational. The device has greatly increased the capabilities and prospects of the laser spectroscopy station at the facility and a new bunched-beam mode of operation has been developed. New results on ground and isomeric states of short-lived and refractory metal isotopes have been reported and further measurements and developments of ultrahigh-sensitivity spectroscopic methods are underway.

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