On-Line Ion Cooling and Bunching for Collinear Laser Spectroscopy

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A new method has been developed for increasing the sensitivity of collinear laser spectroscopy. The method utilizes an ion-trapping technique in which a continuous low-energy ion beam is cooled and accumulated in a linear Paul trap and subsequently released as a short (10–20 μs) bunch. In collinear laser measurements the signal-to-noise ratio has been improved by a factor of 2×10^4, allowing spectroscopic measurements to be made with ion-beam fluxes of $\sim$50 ions s^{-1}. The bunching method has been demonstrated in an on-line isotope shift and hyperfine structure measurement on radioactive ^{175}Hf.

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Laser spectroscopy at on-line isotope-separator (ISOL) facilities has been a key tool in the study of sizes and shapes of exotic nuclei for over twenty years [1,2]. The collinear beams method has been favored in the majority of this work as it eliminates Doppler broadening and makes highly efficient use of the available sample. In the last few years laser-spectroscopic access to short-lived isotopes of a wide range of elements, including the most refractory, has been established at the IGISOL (Ion-Guide Isotope Separator On-Line) facility at the University of Jyväskylä [3,4]. A long-standing problem of applying collinear laser spectroscopy at an ion-guide fed isotope separators is the relatively large energy spread in the ion beam [5–7]. In the early laser work at the IGISOL the energy spread was reduced to a usable level by running the ion guide at a low skimmer voltage [8] but at an unavoidable cost of guide efficiency. The necessary sensitivity was then achieved by using an ion-photon coincidence method to suppress the background [3,9]. Isobaric contamination of the ion beam reduces the sensitivity of the technique and in some mass regions measurements become impossible. In this Letter, we present a new method in collinear laser spectroscopy that removes these limitations and uses an alternative way of suppressing the background. The new method, utilizing ion trapping techniques, has been applied here to a new measurement of the nuclear moments and charge radius of ^{175}Hf.

Ion traps are now appearing as standard tools in manipulating low-energy ion beams. One objective is to improve the emittance and energy spread of the ion beam; another is to convert a continuous beam into short ion bunches [10–13]. The most commonly used ion trap in cooler/buncher applications is the linear variant of the Paul trap [14]. The linear Paul trap was originally developed to reduce second-order Doppler effects in trapped-ion frequency standard applications [15]. The basic principles and construction of the linear trap follow those of the rf-quadrupole mass filter developed by Paul in the 1950's [16,17]. In ion beam manipulation the linear variant is preferred over the traditional Paul trap geometry as there are no high amplitude rf electric fields in the direction of beam propagation, which would, if present, act to prevent the proper matching of a typically continuous ISOL beam into the device.

The ion-beam cooler buncher at the IGISOL facility is positioned immediately downstream of the analyzing magnet of the separator. As such only a desired fraction of the total output of the separator is cooled and unnecessary space charge limitations are avoided. The device consists of a gas-filled rf-quadrupole placed on a high-voltage (HV) platform 100–500 volts below the separator potential that is typically 37–40 kV. The ions are thus decelerated as they enter the cooler and are focused into the quadrupole by two cylindrical electrostatic lenses. The rf-quadrupole is composed of four stainless steel rods of 40 cm in length each divided into 16 segments longitudinally. The applied voltage on an opposite pair of rods is $V = V_{\mathrm{rf}} \cos\Omega t + U_{\mathrm{dc}i}$, where $U_{\mathrm{dc}i}$ is the dc voltage on segment i. Adjacent rods have opposite polarity rf voltage but the same dc voltage. The dc components are used to create a weak electric field in the axial direction towards the exit. The characteristic radius of the quadrupole is $r_0 = 10$ mm. The quadrupole is housed in a buffer gas cell filled with 0.1 mbar of commercial grade 6.0He. The gas cell is "closed" from the injection end by a cylindrical electrode with a 6 mm aperture for the beam injection. At the extraction end there is a miniature quadrupole [18] with identical rf field but smaller radius of $r_{\mathrm{mini}} = 1.5$ mm "closing" the buffer gas volume but keeping the cooled ions still confined in the transverse direction until they are transported to high vacuum. After the miniature quadrupole the ions are accelerated by the HV platform potential and are transported to the experiment. The transmission efficiency and the energy spread of the beam after the cooler have been measured in preliminary nonbunching tests to be 60%–70% and below 1 eV, respectively [11]. A technical description of the construction of the cooler buncher can be found in

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Ref. [11]. The principle of the ion bunching is presented in Fig. 1.

The injected ions are thermalized in the buffer gas and guided with a weak axial field ($\sim 0.1\ \mathrm{V\,cm^{-1}}$) towards the trapping region at the end of the device. In the trapping region a stronger axial field gradient ($\sim 1.2\ \mathrm{V\,cm^{-1}}$) is provided by the last 2 rod segments and the miniature quadrupole. Applying a voltage of $+20$ V on the end-plate electrode creates a potential barrier closing the trap for accumulation of the ions. To release the bunch, the end-plate voltage is switched to zero. The trap serves as a source of cooled ion bunches whose emittance properties are independent of the injected IGISOL beam quality. The ions are accumulated typically for a few hundred milliseconds (t_{acc}). The trap is opened for 100 μs to release the bunch with a width of 10–20 μs. The benefit to collinear laser spectroscopy is diverse. The energy spread of the extracted beam is of the order of 10^{-5} of the total 40 keV energy. At this level the corresponding Doppler broadening of the laser resonance is less than the natural linewidth of most allowed atomic and ionic transitions. The emittance is smaller after the cooler, estimated to be 3π–mm–mrad, resulting in a better spatial overlap between the ion beam and the counterpropagating laser beam. Moreover, the overlap is independent of the ion source and isotope separator, making the experiment insensitive to drifts in the separator or ion guide conditions. Finally, as the ions are in a short bunch of time width w the effective measurement time containing all the real events is a fraction w/t_{acc} of the total measurement time. This allows a suppression of the background events by a factor of $S = (w/t_{\mathrm{acc}})^{-1}$. The efficiency of the suppression depends on the width of the bunch and the collection time that can be used without incurring losses in the ion flux.

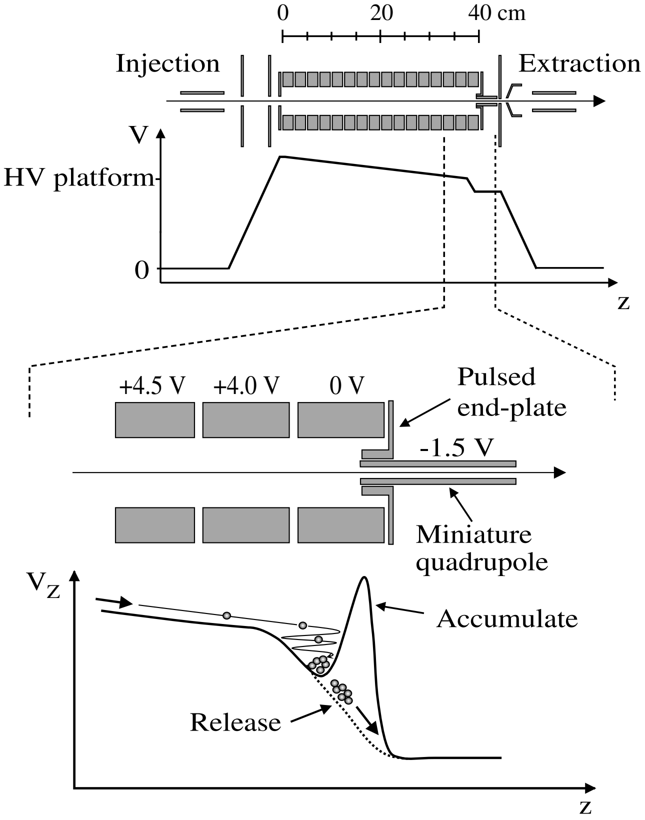

FIG. 1. Schematic principle of the rf-quadrupole ion trap. The ions are accumulated in the trapping zone formed by the rf-quadrupole pseudopotential V_r in the radial direction and by the dc axial potential V_z in the axial direction with pulsed end-plate voltage at +20 V. To release the accumulated bunch, the end-plate voltage is switched to zero leaving the ions in an axial field gradient of $1.2\ \mathrm{V\,cm^{-1}}$ that pushes the ions out of the trap. All the voltages are relative to the HV platform.

The width of the bunch was studied by measuring the ionic resonances of Hf^+. Different gates, synchronized with the ion bunch, were set on photon events from a photomultiplier viewing the ion-laser interaction region. The bunch width was then calculated from the number of resonant photons counted within each gate. Figure 2 shows typical resonance spectra from these measurements. Figure 2(a) shows the photons detected inside a 28 μs gate and Fig. 2(b) shows the total (ungated) photons recorded simultaneously. The photons were simultaneously recorded using two additional gates of 75 and 16 μs. From the relative intensities of resonant photons within these gates the bunch width was evaluated as $w = 14$–16 μs. The bunch width was observed to increase from a minimum value in a manner consistent with a $\sqrt[3]{N}$ dependence where N is the number of ions accumulated in the trap. This can be qualitatively explained, following Ref. [15], by the linear increase of the volume of the ionic ensemble in the harmonic trap (pseudo-) potential as the number of ions is increased. An accurate calculation would be complicated by thermal effects from the buffer

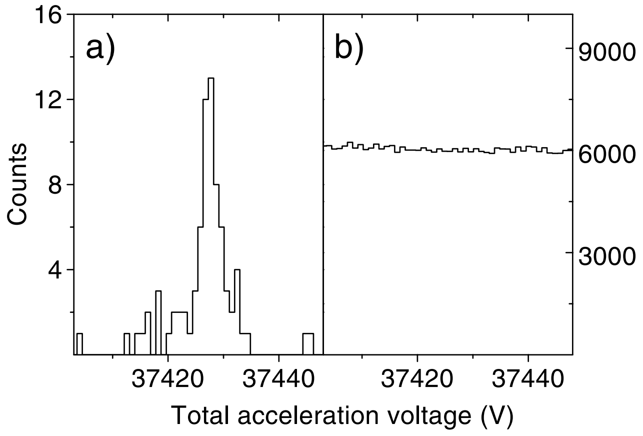

FIG. 2. Resonance fluorescence spectra for $^{174}\mathrm{Hf}$ (a) gated on the ion bunch, and (b) ungated. The spectra show the number of scattered photons counted as the $\mathrm{ds}^2\,{}^2D_{3/2} - \mathrm{dsp}\,{}^2\mathrm{D}_{5/2}$ (301.3 nm) Hf^+ line is brought to resonance with laser light of frequency 33 158.5766 $\mathrm{cm^{-1}}$. The ion rate was 1300 $\mathrm{s^{-1}}$ and the total measurement time was 25 min. The gate used in spectrum (a) was 28 μs and accumulation time in the buncher was $t_{\mathrm{acc}} = 500$ ms.

gas and should be simulated using realistic ion-atom interaction potentials [19]. The increase in the volume of the ionic ensemble restricts the beam intensity that can be efficiently bunched. However, a substantial increase in the bunch width was observed only for $N > 10^4$ below which the width remained approximately constant. With $N = 5 \times 10^4$ the observed width was $w = 18$–$22\ \mu$s. The total mass-separated ion beam intensities from the IGISOL are typically $\sim 10^4$ ions s^{-1} which allows an accumulation time of 1 s before significant space-charge effects are encountered.

The ion survival in the buncher was studied by measuring the resonant photons at constant incident ion beam intensity using different accumulation times. The accumulation time was varied between 100 ms and 1 s. The number of resonant photons per ion injected into the cooler was observed to stay approximately constant independent of the accumulation time. This indicates that either all ions are surviving in the buncher or, perhaps, that losses from the ionic ground state are replaced by other ions relaxing from metastable ionic states (which are known to be populated in the ion source [3]). The spectra in Fig. 2 were collected using an accumulation time of $t_{\mathrm{acc}} = 500$ ms with a beam intensity of 1300 ions s^{-1} and total measurement time of 25 min (30 sec per channel). The background suppression with the 28 μs gate is then $S = 18\,000$. This results in a peak-to-background ratio of 40 at this beam intensity. Extrapolating to lower intensities the new method is applicable to ~ 50 ions s^{-1}. The new technique compares very favorably in spectroscopic efficiency to that achieved with the coincidence system [3] when the latter technique is applied to an isobarically pure beam. Although only measurements on the hafnium system are discussed here, the new technique has been demonstrated to possess comparable off-line efficiencies for other isotope chains including Ti, Y, Zr, Ba, Ce, Yb, Th, and U.

The new method has been used in a measurement of the nuclear moments and charge radius of ^{175}Hf. A previous measurement of the ^{175}Hf charge radius [20] has reported a surprisingly large *normal* odd-even staggering (OES) for this system (with *normal* inferring that the charge radius of ^{175}Hf is less than the average its two neighboring even isotopes). The reported behavior is quite unlike that displayed by odd isotopes in the Yb chain, particularly ^{173}Yb [21], and by other neutron-deficient odd isotopes in the Hf chain [3,22] with these systems displaying a suppression or inversion of the OES in the region around $N = 101$. In the previous, coincidence-based, spectroscopy on hafnium at the IGISOL [3,22] it was not possible to study ^{175}Hf due to a high background ^{175}Lu isobar beam produced from target sputtering inherent in the $^{\mathrm{nat}}\mathrm{Lu}(p, X)$ reaction used. With bunched-beam spectroscopy it has now been possible to investigate the system irrespective of the background ion contamination (provided it is beneath 10^4 ions per bunch). In this work both the $^{175}\mathrm{Lu}(d, 2n)$ and $^{176}\mathrm{Yb}(\alpha, 5n)$ reactions, at 19 and 50 MeV beam energy, respectively, have been used. The Yb reaction was selected for principal study as a 400 μs ^{176m}Hf isomer was abundantly produced by the reaction and could be used to optimize the radioactive output of the ion guide.

The Hf ions were studied on the $\mathrm{ds}^2\,{}^2D_{3/2} - \mathrm{dsp}\,{}^2D_{5/2}$ 301.3 nm line with the ionic ensembles Doppler tuned to resonance with laser light of a fixed frequency. The laser light was provided by a stabilized and frequency doubled spectra physics 380 dye laser with the fundamental laser frequency locked to an I_2 absorption line at $16\,579.2883\ \mathrm{cm}^{-1}$. Fluorescent photons were counted during the period of the ion bunch using the apparatus described in Ref. [22] (without the ion detector or coincidence requirement). Figure 3 shows three scans performed over the ^{175}Hf hyperfine structure.

The best fit to the observed ^{175}Hf hyperfine structure is shown in Fig. 3. The structure was fitted using upper and lower state hyperfine parameters constrained to be in the ratio observed in ^{177,179}Hf with no allowance for hyperfine anomaly. The parameters were determined to be $A(^2D_{3/2}) = -37.4(5)$ MHz and $B(^2D_{3/2}) = +1386(10)$ MHz and the nuclear spin confirmed to be $I = 5/2$. The ^{175}Hf nuclear moments, $\mu = -0.677(9)$ n.m. and $Q_s = +2.72(2)$ b, extracted from these values were scaled from the known nuclear and atomic values of ^{177,179}Hf [3]. Although subject to possible anomaly corrections, the moments for ^{175}Hf are the most accurate available. The values are consistent with most previous reports [20] and fit well into the regional systematics. The isotope shift in the studied transition, $\delta\nu^{175,176} = \nu^{176} - \nu^{175}$, was determined to be $-194(10)$ MHz. Use of the same specific mass shift and field parameters as Levins *et al.* [3] (which follows the calibration obtained by Zimmermann [23]) enables the extraction of the change in mean square charge radius:

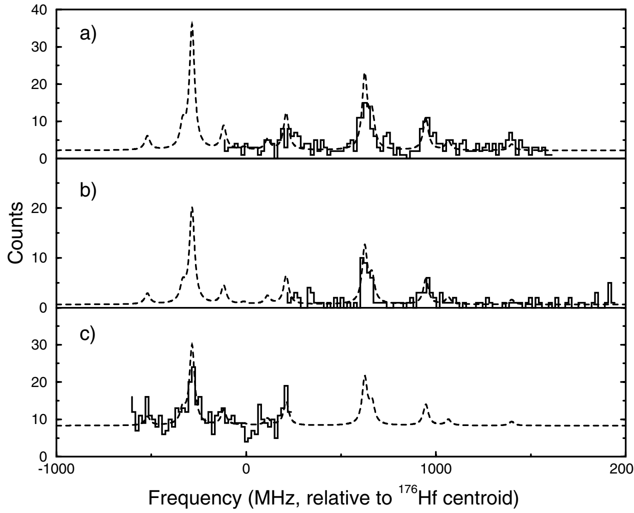

FIG. 3. Resonance fluorescence spectra for ^{175}Hf using, (a), (b) a continuous beam, estimated to contain ~ 2000 ^{175}Hf ions s^{-1}, bunched for 200 ms periods with total scan duration of 100 and 40 min, respectively, and, (c) ~ 400 ^{175}Hf ions $\times\ \mathrm{s}^{-1}$, bunched for 500 ms periods with total scan duration of 400 min.

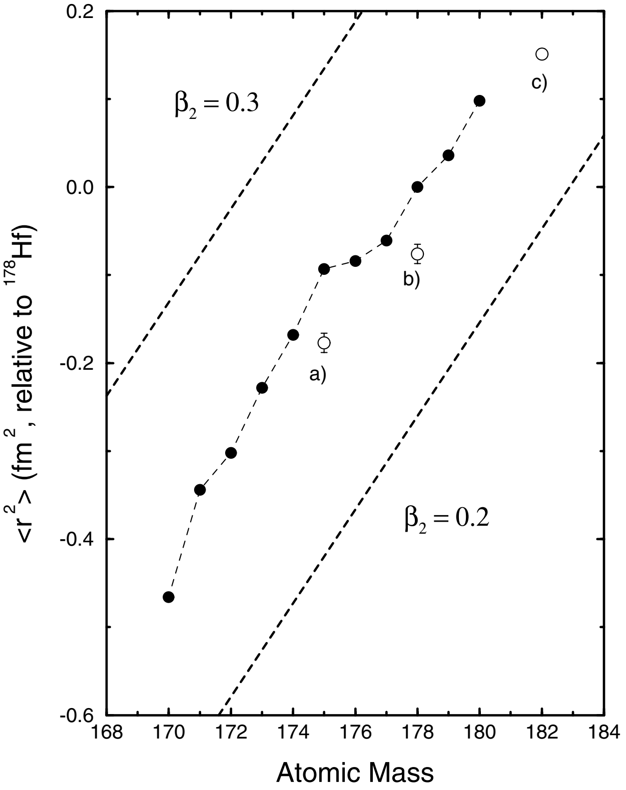

FIG. 4. The hafnium charge radii [22] and new ^{175}Hf measurement [with open circles indicating measurements from other work; (a) ^{175}Hf [20], (b) ^{178m2}Hf [24], and (c) ^{182}Hf [25]]. Deformed droplet isodeformation contours are shown normalized to ^{178}Hf (taking $\beta_2 = 0.252$ for ^{178}Hf).

$$\delta\langle r^2\rangle^{175,178} = +0.093(2)\ \mathrm{fm}^2.$$

The quoted uncertainty reflects only the experimental error and a standard allowance for the specific mass shift uncertainty [3]. An overall calibration error of ~10% [23] is not included. Unlike the nuclear moments, the measured change in mean square charge radius is found to be severely inconsistent with the previous report, $+0.178(11)$ fm^2 [20].

Figure 4 shows the mean square charge radii changes in the hafnium chain. The new measurement shows a large inversion of the OES at ^{175}Hf. Inversions of the OES around $N \sim 101$ are well established in the $Z \sim 80$ region where odd-N isotopes possess far larger quadrupole deformation than their even-N neighbors [1]. This behavior is observed, in this work and Refs. [3,22], to persist down to $Z = 70$. Although far less pronounced than at higher Z, the $N = 99$, 101, and 103 hafnium isotopes display an inverted, or suppressed, OES that is related to their larger static quadrupole moment. The change in charge radius at ^{175}Hf is now entirely compatible with the intrinsic static quadrupole deformation, Q_0 (derived from Q_s), which infers that ^{175}Hf has the most deformed ground state in the hafnium chain. A firm relationship between the course of the radii and changes in the Q_0 moments (estimated from both Q_s and transition rates) has already been established across the rest of the hafnium chain [22], with simple shape and static quadrupole deformation changes appearing to almost exhaustively describe the radial changes of the ground states. The new result for $\delta\langle r^2\rangle^{175,178}$ fits perfectly into this description. Contrary to Jin *et al.* we therefore report a ^{175}Hf charge radius that is almost the same as ^{176}Hf (as opposed to ^{174}Hf). This is compatible with the systematics of the region but no clear explanation for the discrepancy with the previous report can be offered.

Other isotopic and isomeric (particularly high-K) systems in the hafnium chain will be the subject of future investigations with the cooler buncher.

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