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Mass measurements on highly charged radioactive ions, a new approach to high precision with TITAN

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Abstract

TITAN (TRIUMF's Ion Trap for Atomic and Nuclear science) is a system of multiple ion traps installed at the radioactive ion beam facility ISAC. The uniqueness of the system lies in the combination of different kinds of ion traps nowhere else available, and the coupling of this system to ISAC as a source of the most intense radioactive beams of very exotic nuclei worldwide. ISAC is now been operational for more than 5 years, and has been proven to be able to deliver a broad variety of radioactive species with unsurpassed production yields, making it the facility of choice for a next generation ion trap facility, like TITAN.

The physics goals of TITAN are manifold, but the emphasis lies on the test of the Standard Model via the determination of the V_{ud} CKM matrix element, nuclear structure and halo-nuclei investigations, and nuclear astrophysics by providing precise and accurate mass measurements.
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1. Introduction

TITAN consists of five main components; (1) a gas-filled linear radio-frequency quadropole (RFQ) ion trap (RFCT), for cooling and bunching the radioactive beam, (2) an electron beam ion trap (EBIT), for charge-stage boosting, (3) a double Wien-filter for selecting a specific mass-to-charge ratio (WIFI 1&2) (4) a cooler Penning ion trap (CPET), following the Wien-filter, (5) a Penning ion trap for mass determination (MPET) Included are also off-line test ions sources (ISO 1&2) (see Fig. 1.).

The numerous scientific goals of TITAN all share the common characteristic of making use of the unique features of TITAN [1] and ISAC [2]; singly or highly charged, cooled bunched low-energetic beams from the brightest on-line source of radioactive ions. These are prerequisites for high accuracy experiments, since they allow the necessary level of control over the systems, both the system under investigation, here atomic-ions, and the system used to carry out the experiment. High accuracy experiments are planned in the fields of high-precision mass spectroscopy, laser spectroscopy, and x-ray spectroscopy. Motivation for mass measurements is given in the following section.

2. Physics motivation

TITAN will carry out mass measurements of very high precision, with $\Delta m \approx 1$ keV, (or $\delta m/m < 1 \times 10^{-8}$ for $A = 100$ amu), even for isotopes with short half-lives ($T_{1/2} \approx 10$ ms) and for

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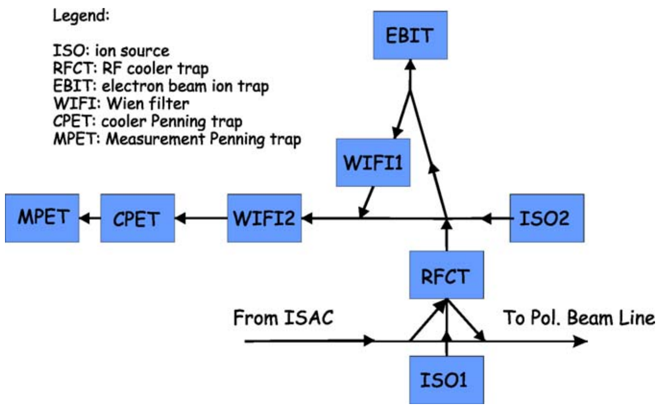

Fig. 1. Schematic layout of TITAN.

isotopes very far from the valley of stability, where production cross sections are small and isotopic yields minuscule. This puts TITAN in a unique situation, where measurements are possible with very high precision on nuclei, that are either not accessible at all for other on-line spectrometers, or cannot be measured with sufficient accuracy. For example, spectrometers such as the Canadian Penning Trap can reach precisions of $\delta m/m \approx 2 \times 10^{-8}$ on isotopes with half-lives of order 1 s. However, the system is coupled to an entirely different production mechanism, and probes in general a different region of the chart of nuclei than is available at ISAC. Spectrometers based on time-of-flight measurements can measure masses of isotopes down to sub-ms half-lives. However, only at the price of accuracies of order $\delta m/m \approx 1 \times 10^{-6}$ or $\Delta m \approx 100$ keV (for $A = 100$ amu). TITAN is therefore ideally suited to address the most demanding questions in the field of mass spectroscopy, particularly regarding tests of the Standard Model of the weak interaction. The Q-value of super-allowed beta-emitters is one of the three required input data for the determination of the FT-value, which together with theoretical corrections leads to V_{ud}, one of the elements of the unitary CKM-Matrix. At present all experimental data, together with the corresponding theoretical corrections, lead to a ≈2 sigma deviation from unitarity [3]. In order to exclude ‘new physics’, the theoretical corrections need to be tested. This can be done in the nuclear physics sector by providing high quality data on other beta-decaying nuclei, where the same theoretical framework is applied but which would be more sensitive to errors in the corrections.

In particular, the recent determination of the Q_{EC} value for decay of the superallowed (-emitter ^{46}V obtained by measuring the difference in the masses of ^{46}V and its decay daughter ^{46}Ti with the CPT [4], not only improved the precision of this quantity but also revealed a systematic error in a set of 7 measurements that affected previous evaluations of V_{ud} by 0.02%. This clearly demonstrates the need for high-accuracy data of new and previously measured nuclei.

3. Principle and status of individual components

The individual components of TITAN can be operated as stand alone units, and optimized in that way using the off-line sources. The beam will ultimately be transferred from the ISAC beam line through the RFCT to the EBIT, and via the two Wien filters to the cooler Penning trap and the measurement Penning trap. The transfer is critical and has to be highly efficient. The transfer system is designed using the standard low-energy beam-line components of ISAC and a higher order transfer code [5]. For the ISAC beam line, the transfer efficiency is on average better than 90%. In the following the main components are described in detail.

3.1. Buffer-gas filled radio frequency quadropole cooler and buncher trap (RFCT)

The RFCT is the first component of the TITAN facility. The function of the RFCT is to take the continuous 30–60 keV ISAC beam, cool it via interactions with a neutral buffer gas, and convert it to a bunched beam of variable energy of up to 5 keV. From simulations, injection efficiencies into the RFQ after deceleration of close to 100% were expected and around 90% has been achieved. The ions can only be trapped if they enter the RFQ structure with about 50 eV. Hence all excess kinetic energy has to be removed by floating the RFQ structure on a high voltage (HV) platform. This was successfully demonstrated to work at up to voltages of 65 kV. The cooling mechanism with the neutral buffer gas leads in general to an increase in beam size, which is countered by the RF fields. The cooling process and ion transport were simulated with a newly developed Monte Carlo code, where experimental data for ion–atom collision potentials were used. The results of these simulations can be compared to the experimental results.

In order to quantify the effectiveness of the cooling process, and to find optimal parameters, an emittance meter was designed and constructed. This device allows measurements on weak (pA) bunched beams. The results agree well with the simulations and show a transversal emittance of $\varepsilon \approx 6\pi$ mm mrad at 5 keV extraction energy (Fig. 2), corresponding to an improvement of over a factor of 30.

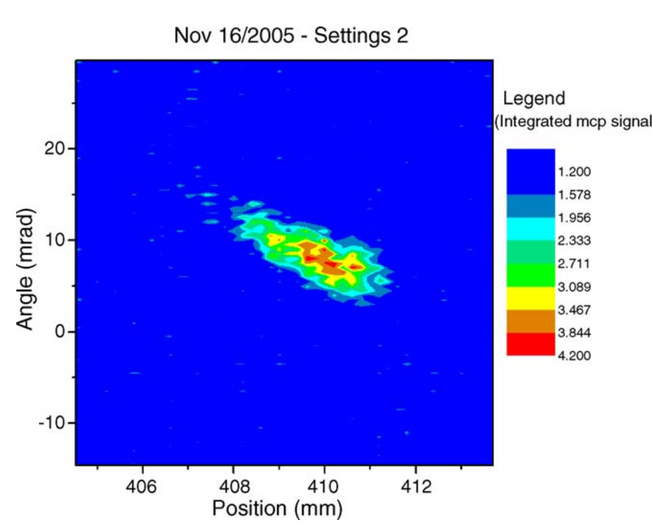

Fig. 2. Emittance plot of the extracted beam from the buncher.

A novel aspect of the TITAN RFQ, compared to other cooler and buncher devices operational at RIB facilities, is the way the RF fields are generated. The challenge for the driver is that stable trajectories of the ions can be achieved with the proper combination of RF frequency and amplitude, physical dimensions of the RFQ structure and of course the mass-to-charge ratio of the ions. The latter can vary significantly, for example, by a factor of almost 30 between ^{8}Li to ^{224}Fr, which has to be accommodated by varying the frequency and/or the amplitude. The latter should stay as high as possible, since the amplitude is directly proportional to the space charge limit in the RFQ. If the buncher is to accept the ISAC beam completely a high space charge limit is desired. The RF driver approach for the TITAN cooler and buncher is therefore based on hardware that allows one to provide a high amplitude RF field over a broad frequency band, from 300 kHz to 3 MHz. The RF is generated by pairing an up and a down MOSFET switch, generating a square wave. Two such systems are then used for opposite phases. Moreover, the switches can be stacked, resulting in an adding of the potential difference, hence increasing the amplitude. Presently a system that is capable of providing up to 1000 V_{pp} is used, but can be extended to high amplitudes. The RF system is working reliably and is controlled via an optical link from the VME-based control system EPICS on ground potential [6]. The estimated space charge limit of the buncher in continuous mode is around 2 (A in DC operation, hence without bunching, or 32 nA with a 1 ms cooling-time bunching cycle with our present system. This is clearly sufficient for beams of short-lived isotopes produced on-line at ISAC.

3.2. Electron beam ion trap (EBIT)

The electron beam ion trap is the second main component for TITAN. The purpose is to boost the charge state of the singly charged ions delivered from ISAC by rapid charge breeding. The process has to be both fast and efficient so as to accommodate the stringent requirements when operating with very short-lived ions. The novel TITAN approach to this challenge is an EBIT [7] with very high electron current densities. The device is currently assembled and tested in collaboration with the Max-Planck Institute (MPI) for Nuclear Physics in Heidelberg.

The basic principle of an EBIT is to employ an electron beam to provide a negative space charge potential in which the positively charged ions are trapped radially. Additional electrodes provide longitudinal confinement. The electron beam will then strip away shell electrons from the ions. If the stripping is fast enough, and if the electron energy is higher than the atomic binding energy, higher charge states are produced. The breeding time is inversely proportional to the electron charge density. The latter is increased significantly by the compression in a 6 T magnetic field. Theoretical modeling has shown to reach for example Rb^{35+} (He-like) in 30 ms.

Fig. 3 shows a schematic overview of the TITAN-EBIT. The radionuclides enter the EBIT as cooled bunches of singly charged ions through the collector. The extraction after charge breeding takes place along the same path in the opposite direction. The extraction and the transport to the precision Penning trap will be accomplished by means of floatable drift tubes and pulsed cavities. In June 2005, a first setup of the TITAN-EBIT was assembled. With the prototype version of the trap and the electron gun and collector system still at ground potential, the creation of electron beam currents of up to 190 mA and kinetic energies of 14 keV were achieved. Besides trapping and ionizing, the electron beam gives rise to excitations of the trapped ions and recombination. A germanium detector was placed at one of the seven radial ports to observe x-rays from the trap region through a beryllium vacuum-window. A sample X-ray spectrum taken during the first week of operation is shown in Fig. 4. Barium (and tungsten) that is evaporated from the cathode surface makes its way as neutral atoms into the trapping region where it gets ionized to high charge states. The spectrum

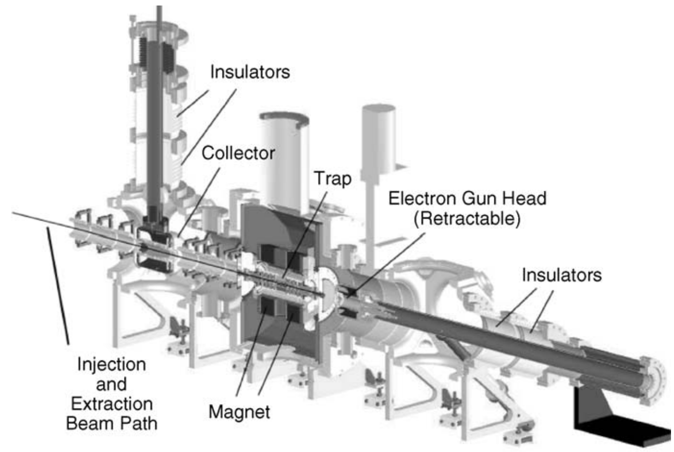

Fig. 3. Schematic overview of the TITAN-EBIT. A superconducting 6 T magnet with cold bore houses the actual trap setup. Both, the electron gun head and the electron collector unit are adjustable with respect to the magnetic field and relative to each other to provide an optimal electron beam performance. The electron gun and the collector are floating on negative high voltage whereas the trap is held at ground potential.

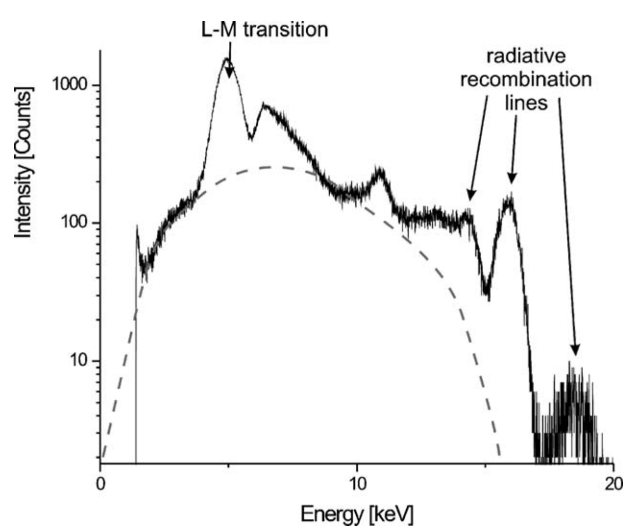

Fig. 4. X-ray spectrum from trapped highly charged Ba ions. Spectral lines can be seen on top of the bremsstrahlungs-continuum (dashed line). Besides direct transitions, of which the L–M transition is the most prominent one, resonances at energies much higher than the kinetic energy (16.5 keV) of the electrons are observed.

gives clear evidence for trapping and ionization of Ba. The most prominent excitation and recombination lines were observed. This detection technique will be available as a diagnostic tool during on-line operation.

At the present time, the final version of the trap, including a gas injection system is being implemented. The latter will allow one to increase the variety of elements for tests and to provide for insertion of cooling gases such as argon or neon. The operation of the complete TITAN-EBIT will start in December 2005. First measurements will focus on the creation of highly charged ions from neutral gas atoms and their extraction. The main focus will lie on the distribution of charge states as a function of the primary EBIT parameters, such as the electron energy and current density. Recent studies of the EBIT group demonstrated a new method to maximize the fraction of ions in a single charge state by varying the electron beam energy [8]. This yielded an increase of the charge state of interest (here Li-like Kr) of factor of 2–4. Moreover, buffer gas cooling was investigated at the same EBIT in Heidelberg and energy spreads of 6 eV/q [9] could be reached. At RexEBIS [10], where injection and extraction of short-lived ions has been studied in detail, ratios of injected-to-extracted ions of close to one have been reached. Similar values are expected for the TITAN system.

3.3. Cooler Penning trap (CPET)

Little work has been done so far in characterizing the emittance of beams extracted from EBITs. From the available information (Livermore [11], RIKEN [12], Dresden EBIT [13], and REX-EBIS [10]), it is apparent that energy spreads of the order of many eV/q must be expected. Generally, precision mass measurement in a Penning trap requires ion temperatures of $kT_i \leq 1$ eV/q. The push for very high electron currents of up to 5 A in the TITAN EBIT will most likely worsen this problem. For this reason, we plan to insert a cooler Penning trap between the charge breeding and the mass measurement. For highly charged ions, this poses new challenges. The method of buffer gas cooling, used very successfully with singly or doubly charged ions, is ruled out by the unacceptably large charge transfer; resistive cooling requires a cryogenic environment and is highly q/m specific, making it more suitable for dedicated work with a particular species. The most promising general purpose route to cooling of HCIs at this point is to use sympathetic cooling via the Coulomb interaction with a cold ensemble of non-ionizable, charged particles such as electrons, positrons, or protons.

Electron cooling is attractive as electrons are easily produced and self-cool to the temperature of the environment via the emission of synchrotron radiation with a time constant of ≈100 ms in a 6 T magnetic field. A complication is the occurrence of electron-ion recombination. For typical parameters such as an electron temperature of 300 K, an electron density of 10^7 cm^{-3}, and $q = 50$, the radiative recombination rate amounts to ≈0.1 s^{-1}, once the ions are at energies below ≈100 eV/q. This is quite tolerable. However, higher electron densities (desirable for faster cooling), dielectronic recombination, and three-body recombination could shorten the lifetime significantly. In addition, the opposite charge of electrons and ions requires the use of a *nested* trap, where electrons are trapped in one or multiple inverted wells inside a larger ion trap. At ion energies corresponding to the height of the inverted well, the two species start to decouple spatially, halting the cooling process.

Positron cooling would avoid both the recombination and the decoupling, but adds significant technical complication and cost, as it requires a positron source at the 100 mCi level. Nevertheless, positron cooling of highly charged ions has been successfully demonstrated by Oshima et al. at RIKEN. At a later stage, it might be worth considering.

As an alternative, we are looking into light-ion cooling of highly charged ions, where sub-eV protons from an ion source are injected into the trap. The synchrotron self-cooling rate scales with m^{-3} and hence this mechanism cannot be relied upon for protons, hence a ‘cold’ proton source is required. Initial test have shown that protons with low energy spread are available. Calculations indicate that this method can cool highly charged ions at time scales much shorter than 1 s [14], potentially as short as 10 ms. At present the system is designed and will be set up and tested at the University of Manitoba. After commissioning the CPET will be included in the TITAN set-up.

3.4. Mass measurement Penning trap (MPET)

The core piece of the TITAN setup is the Penning trap mass spectrometer. Penning traps are the most precise devices to measure masses and our system is designed to carry out measurements of atomic masses with an accuracy of $\delta m/m < 1 \times 10^{-8}$, even for radioactive isotopes with half-lives well below 100 ms. The resolving power of this measurement method is proportional to the inverse of the excitation time T_{RF}

$$\frac{m}{\delta m} \propto \omega_c T_{RF}\sqrt{N} = \frac{qB}{m} T_{RF}\sqrt{N}$$

where N is the statistical improvement factor due to tracing the resonant curve N times. Ideally one would try to increase the excitation time as much as possible. However, in the case of stable ions, this is limited by the storage time in the trap, and for

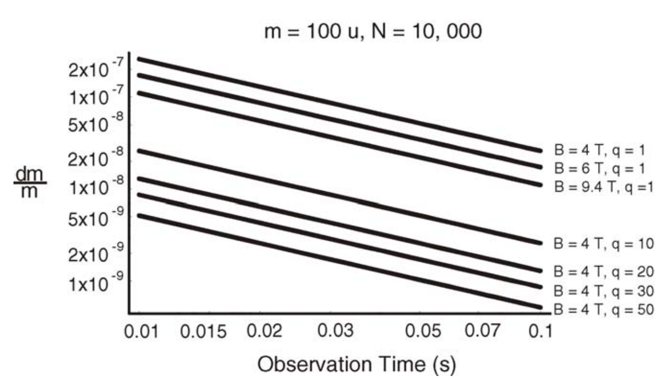

Fig. 5. Relative accuracy of the Penning trap measurement as a function of observation time (typically two nuclear half-lives correspond to the applicable observation time). The different sets of graphs represent different charge states q and different magnetic field strength B. The case shown is for 10 000 measurement cycles and ions with mass 100.

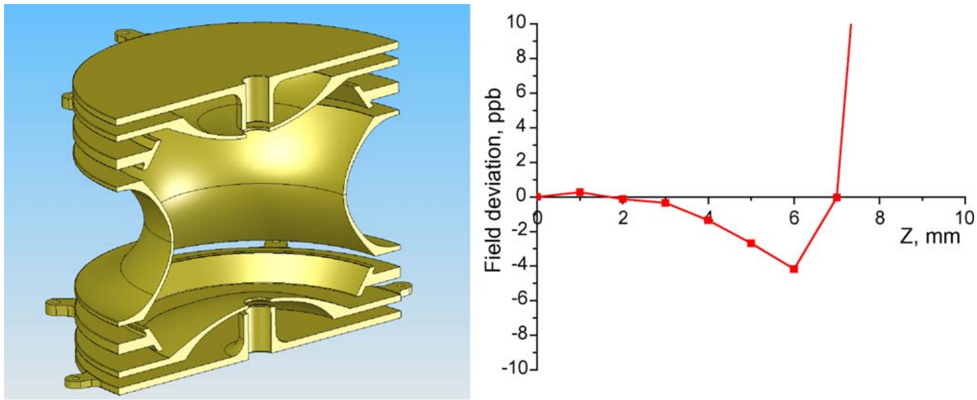

Fig. 6. Section view of a rendered drawing of the electrodes of the precision Penning trap (left). (b) disturbance to the magnetic field by the Penning trap electrodes (right).

the case of short-lived isotopes it is limited by the decay half-life of the ion. Similarly, increasing the magnetic field will give an increase in precision, yet the improvement is limited by the current magnet technology. The TITAN setup will be the first online system worldwide to utilize the significant increase in the charge q of the measured ion to improve the accuracy.

Fig. 5 shows the relative accuracy of Penning trap spectrometers as a function of measurement time, in sets of magnetic field strength B and charge state q of the ions. The TITAN system with a magnetic field of 4 T will allow measurements with accuracies of better than $\delta m/m < 1 \times 10^{-8}$ on isotopes with half-lives as short as 20 ms, when ions with charge states of $q = 20$ are used.

The electric and magnetic field homogeneity in the measurement Penning trap is crucial for achieving the desired accuracy. Therefore, a series of electrostatics calculations were performed to achieve the best electric field parameters. Moreover, the material along the trap electrodes was redistributed by successive optimizations to obtain the best possible magnetic field homogeneity. The magnetic field calculations were incorporated into the CAD design program. The resulting trap geometry and the corresponding magnetic field inhomogeneity are shown in Fig. 6.

The present design will lead to field perturbations in the region of interest of less than 2 ppb. Hence, mass measurements of the desired accuracy of $\delta m/m < 1 \times 10^{-8}$ are possible. Another challenge for the TITAN Penning trap system is the requirement of ultra-high vacuum, which is needed to reduce the collisions of the highly charged ions with residual background gas. The latter would lead to unwanted charge exchange processes, and therefore to losses. The requirements are to reach a pressure of $p < 5 \times 10^{-10}$ mbar. At this level the trapping times are expected to be on order of seconds. The complete vacuum system is designed and all components are ordered. The magnet is scheduled for delivery in early 2006 and first tests are planned for the summer of 2006.

4. Expected performance

The successful mass determination of short-lived isotopes requires am efficient and fast preparation of the ions prior to their loading into the measurement Penning trap. The expected efficiencies and processing times are giving in Table 1. For the charge breeding process a moderate charge state (for example Li-like) is assumed, where a balance between breeding time and gain in precision can be reached.

Table 1
Expected efficiency and processing time for the different components of TITAN.

Component	Expected efficiency	Processing time (ms)
RFCT	0.7	2
EBIT	0.5	5
EBIT charge state	0.3	
CPET	0.5	10 (up to 100)
Transfer beam line	0.1	
SUM	0.00525	17 (up to 107)

The total efficiency to prepare the sample and transfer it to the MPET is around 0.5% with an expected processing time of around 20 ms. The processing time is dominated by the cooling time in the CPET and is strongly depending on the achievable parameters in the cooling process, in particular the reachable densities of the cold ions have a direct influence on the cooling time. For an assumed processing time of 20 ms (110 ms) decay losses have to be taken into account. The mass measurement itself requires an excitation time on the order of 2 half-lives, which contributes to the total time. Hence a total of 3 half-lives for isotopes with $T_{1/2} = 20$ ms and an overall efficiency of 0.5% would require a minimal yield of around 1000 ions/s to perform experiments on the level of $\delta m/m < 1 \times 10^{-8}$. This would be significantly altered if much longer cooling times in the CPET, in contrast to theoretical predictions, would be required. For longer-lived isotopes, lower yields would be required, for example for isotopes with $T_{1/2} = 100$ ms around 300 ions/s would be sufficient.

5. Conclusions

Mass measurement devices for exotic nuclei exist at various places and in different forms. The apparatus proven to be the most accurate system, the Penning trap, is becoming more and more popular, and there exist 5 systems that are presently coupled to on-line facilities, ISOLTRAP (ISOLDE, CERN), CPT (ATLAS, ANL), SHIPTRAP (GSI), JYFLTRAP (Jyväskylä), and LEBIT (NSCL/MSU). All systems are by now operational and produce experimental results, at various levels of precision. The systems are in general complementary to each other, since they are specifically built to fill the need for experiments in a particular niche of nuclear physics. They are different either in the way the radio-isotopes are produced (ISOL, fragment separator, IGISOL, reaction & gas cell), or the way the spectrometer is setup.

TITAN will be the only on-line spectrometer to employ highly charged ions, and hence will have the capability to reach a level of accuracy that is unattainable elsewhere. At present, the highest accuracy required by experiments (for the V_{ud}-determination) appears to be $\delta m/m \approx 1 \times 10^{-8}$. However, now other possible experiments, are being considered, for example, $2\beta0\nu$ experiments, where the decay matrix element determination requires a knowledge of the Q-value to the eV level, or $\delta m/m \approx 1 \times 10^{-9}$. With further developments TITAN will be able to carry out such experiments.

The utilization of highly charged ions, and the superior production yields from the production method at ISAC, where the world's highest ISOL primary driver power is available to produce exotic species puts TITAN in a unique position for experiments of unprecedented accuracy and on isotopes nowhere else available with the required intensities. The construction and set-up phase are well underway and first on-line experiments are planned for 2006.

Acknowledgments

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References

[1] TITAN collaboration, Nucl. Instrum. Methods B 204 (2003) 492.
[2] M. Dombsky, P. Bricault, T. Hodges, A. Hurst, P. Schmor, Nucl. Phys. A. 701 (2002) 486.
[3] J.C. Hardy, I.S. Towner, Phys. Rev. Lett. 94 (2005) 92502.
[4] G. Savard, F. Buchinger, J.A. Clark, J.E. Crawford, S. Gulick, J.C. Hardy, A.A. Hecht, J.K.P. Lee, A.F. Levand, N.D. Scielzo, H. Sharma, K.S. Sharma, I. Tanihata, A.C.C. Villari, Y. Wang, Phys. Rev. Lett. 95 (2005) 10250.
[5] R. Baartman, Nucl. Instrum. Methods B 204 (2003) 392.
[6] M.J. Barnes, G.D. Wait, J. Dilling, J.V. Vaz, L. Blomeley, O. Hadary, M.J. Smith, Proceedings of the IEEE International Pulsed Power Conference San Fransisco, Ca, USA, 2005.
[7] G. Sikler, J.R. Crespo López-Urrutia, J. Dilling, S. Epp, C.J. Osborne, J. Ullrich, Eur. Phys. J. A 25 (2005) 1.63.
[8] J.R. Crespo López-Urrutia, J. Braun, G. Brenner, H. Bruhns, A. Lapierre, A.J. González Martínez, V. Mironov, R. Soria Orts, H. Tawara, M. Trinczek, J. Ullrich, Rev. Sci. Instrum. 75 (2004) 1560.
[9] R. Soria, PhD Thesis University of Frankfurt 2005.
[10] F. Wenander, Nucl. Phys. A 746 (2004) 40.
[11] B.M. Penetrante, J.N. Bardsley, Phys. Rev. A 43 (1991) 4861.
[12] N. Oshima, T.M. Kojima, M. Niigaki, A. Mohri, K. Komaki, Y. Iwai, Y. Yamazaki, Nucl. Instrum. Methods B 205 (2003) 178.
[13] U. Kentsch, G. Zschornack, F. Grossmann, V.P. Ovsyannikov, F. Ullmann, Nucl. Instrum. Methods B 205 (2003) 266.
[14] V. Ryjkov, et al., Eur. Phys. J. A 25 (2005) 1.53.