High precision Penning trap mass spectrometry of rare isotopes produced by projectile fragmentation

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Abstract. The Low Energy Beam and Ion Trap (LEBIT) is the only present facility to combine high precision Penning trap mass spectrometry with fast beam projectile fragmentation. Located at the National Superconducting Cyclotron Laboratory (NSCL), LEBIT is able to measure radionuclides produced in a chemically independent process with minimal decay losses. Recent exotic mass measurements include 66As, 63−66Fe, and 32Si. 66As is a new candidate to test the Conserved Vector Current (CVC) hypothesis. The masses of the neutron-rich iron isotopes provide additional information about the mass surface and the subshell closure at N = 40. 32Si is a member of the A = 32, T = 2 quintet; its measurement permits the most stringent test of the validity of the isobaric multiplet mass equation (IMME). An overview of some recent measurements will be presented as well as advanced techniques for ion manipulation.

1. Introduction
Mass measurements provide insight into many nuclear physics topics such as element synthesis in supernovae, nuclear shell and subshell closures, and the search for scalar currents. The relative precision required for these applications ranges from 10−5, e.g. for observing the onset of nuclear deformation, to better than 10−8 for tests of the Conserved Vector Current (CVC) hypothesis [1]. The production of rare isotopes by fast beam projectile fragmentation is attractive for a variety of reasons. Since chemical effects are avoided and decay losses are minimal, a large number of nuclides can be created and harvested ranging from the valley of stability to the proton- and neutron-driplines. The development of gas thermalization techniques permit low energy, precision experiments to take advantage of these exotic and short-lived nuclei, which may be difficult or even impossible to produce with other techniques. The Low Energy Beam and Ion Trap (LEBIT) facility at Michigan State University was the first to perform high precision mass spectrometry on thermalized projectile fragment products.

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2. Experimental Setup
The LEBIT facility consists of a high pressure gas stopping station to thermalize the beam, a two stage cooler and buncher to cool and to bunch the ions, and the 9.4 T Penning trap mass spectrometer for high precision mass measurements.

2.1. The Gas stopping station
Projectile fragments are delivered to the linear gas cell with energies on the order of 100 MeV/u. The beam passes through an adjustable glass degrader system, where most of its kinetic energy is dissipated, through a thin beryllium entrance window, and then into the 51 cm long linear gas cell [2, 3]. The ions are stopped in high purity helium gas at a pressure up to one bar. An electrostatic field guides the ions towards the nozzle, where gas flow ejects the ions from the gas cell. A series of three radio frequency quadrupole (RFQ) ion guides transport the beam through a differential pumping system into high vacuum.

The helium gas only has impurities on the level of a few ppb; nonetheless, as the helium ions created during the stopping process spend tens of milliseconds in the gas cell, they can exchange charge with residual impurities and thereby form molecular ions. The last section of the RFQ ion guides is operated as a mass filter [4] to separate the ion of interest from the stable contaminant ions before its being transported to the cooler and buncher.

2.2. The Cooler and buncher
The cooler and buncher converts the continuous ion beam into short pulses with a small transverse and axial emittance [5]. The first or “pre-cooler” stage, an RFQ ion guide filled with a buffer gas at \( \approx 10^{-2} \) mbar, is where the ions are slowed and their transverse emittance is reduced. In the second stage, a linear Paul trap filled with helium buffer gas at \( \approx 10^{-4} \) mbar, ions are accumulated and cooled to thermal equilibrium before they are ejected as an ion pulse \( \approx 100 \) ns long. The small time spread permits additional mass filtering by using a fast pulsed beam gate to select ions on their way to the Penning trap with single mass resolving power.

Since the gas pressures in the first and second stages are largely decoupled, different buffer gases may be used in each stage. Helium gas is typically used in both sections; however, neon may be introduced into the pre-cooler to separate radioactive species from isobaric stable molecular contaminants via collision-induced dissociation [4]. The energetic molecular ion (>10 eV) collides with the buffer gas and generally emits a neutral fragment leaving behind a molecular ion with a very different mass-to-charge ratio.

![Figure 1. Mean time-of-flight as a function of the RF frequency in the Penning trap, \( \nu_{RF} \) for \( ^{32}\text{SiOH(H}_2\text{O)}^+ \). The solid line is a fit of the theoretical line shape [6] to data.](image-url)
2.3. The Penning trap system

The LEBIT Penning trap [7] is a high precision hyperbolic electrode system at the center of an actively shielded 9.4 T superconducting magnet. There are three eigenmotions in the trap: reduced cyclotron motion $\nu_+$, magnetron motion $\nu_-$, and axial oscillation $\nu_z$ [8, 6]. The true cyclotron frequency $\nu_c$ and, hence, the mass can be determined since in a perfect trap

$$\nu_c = \nu_+ + \nu_- = \frac{1}{2\pi} \frac{q}{m} B \tag{1}$$

where $q$ and $m$ are the charge and mass of the ion and $B$ is the magnetic field strength. At the LEBIT facility, the time-of-flight (TOF) resonance detection technique is used [6, 9]. The ion pulses are captured in the trap, exposed to a radiofrequency (RF) field at a frequency $\nu_{RF}$ close to $\nu_c$, and ejected from the trap. The time between ejection and the ions striking an MCP detector located outside of the magnetic field is recorded. When the appropriate excitation time and RF amplitude are used and $\nu_{RF} = \nu_c$, the initial magnetron motion is fully converted into cyclotron motion, increasing the radial energy. This radial energy is converted into axial kinetic energy as the ion traverses the magnetic field gradient at the end of the magnet and registers as a shorter TOF to the MCP. Repeated cycles of capture, excitation, and TOF measurement produce a resonance centered at $\nu_{RF} = \nu_c$, like the one shown in Figure 1 for $^{32}\text{SiOH}(\text{H}_2\text{O})^+$. To calibrate the magnetic field, typically the cyclotron frequency of a stable molecular reference ion with a well known mass is measured before and after the resonance frequency of the ion of interest is measured. The magnetic field strength is obtained by linear interpolation between the two calibration measurements. At a field strength of 9.4 T, the same precision can be reached in half the excitation time required in more common 6 T systems. To reduce the measurement time further, a Lorentz steerer [10] is used to place the ions in the trap with a particular magnetron radius.

3. Mass Measurements

Since the initial operation of LEBIT in 2005, the masses of more than 11 elements and approximately 40 isotopes have been measured at the LEBIT facility with relative uncertainties

![Figure 2. The two neutron separation energy $S_{2n}$ as a function of the neutron number $N$. The red circles correspond to LEBIT data. Gray circles represent data taken from AME'03 [11] and more recent data [12]. Filled circles are calculated from measurements, open circles from extrapolations based on systematic trends [11].](image-url)
as low as 10 ppb [13, 14, 15]. The latest measured mass is that of $^{66}$As [16], which is a superallowed $\beta$-emitter, and with an uncertainty now reduced to $\approx$6 keV, it is a candidate to test the Conserved Vector Current (CVC) hypothesis. Other recent measurements have been performed for nuclear structure studies of neutron-rich sulfur up to the $N = 28$ subshell closure [17], of neutron-rich iron and cobalt isotopes [18, 19], and of $^{32}$Si to test the Isobaric Multiplet Mass Equation (IMME) [15]. The last two cases will be discussed below in greater detail.

3.1. Neutron-rich Fe and Co isotopes

The structure of nuclei in the $N \approx 40, Z < 28$ region has some interesting features. While the $E_x(2^+)$ and the $B(E2)$ value of $^{68}$Ni indicate a strong subshell closure at $N = 40$ [20], $\beta$-spectroscopy of $^{66}$Fe [21] suggests deformation, and theoretical calculations of neutron-rich iron isotopes [22] suggest shape coexistence. The LEBIT campaign of mass measurements of $^{63-66}$Fe and $^{64-67}$Co [18, 19] complements earlier studies. From examining the two neutron separation energies, shown in Figure 2, it appears that the $N = 40$ subshell closure occurs very weakly in cobalt similar to higher $Z$ nuclei [23, 24]; however, in iron the closure appears to vanish. Further measurements to reduce the mass uncertainties in Fe isotopes beyond $N = 40$ should be able to confirm the vanishing subshell closure. During the measurement of the mass of $^{65}$Fe, a second resonance was observed. It corresponds to a previously unknown, long-lived ($T_{1/2} \gtrsim 150$ ms) isomer in $^{65}$Fe.

3.2. The Isobaric multiplet mass equation

The Isobaric Multiplet Mass Equation (IMME) [29] is used to describe the masses of isobaric analog states with a quadratic function of the isospin projection $T_z = (N - Z)/2$. It is determined from first-order perturbation theory under the assumption that the charge-dependent perturbations arise from pair interactions. The $A = 32, T = 2$ quintet was considered the most stringent test case of IMME [30]: recent measurements of the ground states and first $T = 2$ excited state energies [31, 32, 25] first upheld and later invalidated IMME. Discussion had focused on isospin mixing in $^{32}$S or an erroneous mass for $^{32}$Si [25]; an analysis of the data indicated that if $^{32}$Si were 3 keV more bound, the quadratic form of IMME would be restored. The LEBIT mass measurement of $^{32}$Si found a deviation of 3 keV in the direction opposite that needed to restore IMME; indeed, the deviation served to amplify the breakdown of the assumed quadratic form of IMME. More recently, the mass of the first $T = 2$ state in $^{32}$Cl has
been remeasured [26] and confirmed the need for a non-zero cubic term [26]. Non-zero cubic or quartet terms may result from higher order pure Coulomb terms, isospin mixing in the first \( T = 2 \) excited states of \(^{32}\text{P}, ^{32}\text{S}, \) and \(^{32}\text{Cl}\), or other charge-dependent effects. A cubic coefficient up to \( \approx 1 \) keV has been predicted using simple models [33, 34]; however, to date no calculations specific to the \( A = 32, T = 2 \) quintet have been published. The results of a fit to the quadratic and cubic forms of IMME are shown in Figure 3.

4. Summary and outlook
The mass measurement campaign at the LEBIT facility has made contributions to a variety of topics in nuclear physics, ranging from fundamental symmetries to shell closures in nuclear structure. Many of the isotopes measured at LEBIT are difficult or even impossible to measure at other Penning trap mass spectrometers that rely on ISOL beam production.

While fast beam fragmentation offers chemistry-independent and fast isotope delivery, unforeseen challenges to thermalize and to purify the beam have arisen since LEBIT first began operation. The gas stopping station at the NSCL is undergoing several major upgrades to improve extraction efficiency, to reduce extraction times, and to improve beam purity. Recently, the LEBIT system has been decommissioned and relocated to a new experimental vault. As part of the recommissioning, several developments are being pursued including SWIFT (Stored Waveform Inverse Fourier Transform) for in-trap beam purification and a miniature Penning trap to be used as a magnetometer. Both developments will optimize the use of beam time and reduce possible sources of systematic errors.

Acknowledgments
The authors thank those who provided support throughout the project: C. Bachelet, M. Facina, C.M. Folden III, A.E. Gehring, C. Guénaü, A.M. Prinke, D. Melconian, and S. K.J Sjue. This work has been supported by Michigan State University, the National Science Foundation under contract number PHY-0606007, and in part by the Department of Energy under contract number DE-FG02-00ER4114.

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